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RECOMMENDED METHODS OF REDUCTION, NEUTRALIZATION, RECOVERY, OR DISPOSAL OF HAZARDOUS WASTE

VOLUME IX. NATIONAL DISPOSAL SITE CANDIDATE WASTE STREAM CONSTITUENT PROFILE REPORTS. RADIOACTIVE MATERIALS

TRW Systems Group

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This volume contains summary information and evaluation of waste management methods in the form of Profile Reports for radioactive materials. These Profile Reports were prepared for either a particular hazardous waste stream constituent or a group of related constituents. Each Profile Report contains a discussion of the general characteristics of the waste stream constituents, their toxicology and other associated hazards, the definition of adequate management for the waste material, an evaluation of the current waste management practices with regard to their adequacy, and recommendation as to the most appropriate processing methods available and whether the waste material should be considered as a candidate for National Disposal, Industrial Disposal, or Municipal Disposal.

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National Disposal Site Candidate
Hazardous Wastes
Carbon-14
Cobalt-60
Iridium-192
Radium-226
Cesium-134
Cesium-137
Hydrogen-3
Iodine-129
Iodine-131
Krypton-85
Xenon-133
Plutonium-238, 239, 240, 241
Amercium-241, 243
Curium-242, 244
Ruthenium-106
Cerium-144
Promethium-147
Strontium-90
Zirconium-95
Niobium-95

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Volume IX. National Disposal Site Candidate Waste Stream Constituent Profile Reports - Radioactive Materials

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FOREWORD

Man and his environment must be protected from the adverse effects of pesticides, radiation, noise and other forms of pollution, and the unwise management of solid waste. Efforts to protect the environment require a focus that recognizes the interplay between the components of our physical environment--air, water, and land. The National Environmental Research Centers provide this multidisciplinary focus through programs engaged in:

- studies on the effects of environmental contaminants on man and the biosphere, and
- a search for ways to prevent contamination and to recycle valuable resources.

Under Section 212 of Public Law 91-512, the Resource Recovery Act of 1970, the U.S. Environmental Protection Agency is charged with preparing a comprehensive report and plan for the creation of a system of National Disposal Sites for the storage and disposal of hazardous wastes. The overall program is being directed jointly by the Solid and Hazardous Waste Research Laboratory, Office of Research and Development, National Environmental Research Center, Cincinnati, and the Office of Solid Waste Management Programs, Office of Hazardous Materials Control. Section 212 mandates, in part, that recommended methods of reduction, neutralization, recovery, or disposal of the materials be determined. This determination effort has been completed and prepared into this 16-volume study. The 16 volumes consist of profile reports summarizing the definition of adequate waste management and evaluation of waste management practices for over 500 hazardous materials. In addition to summarizing the definition and evaluation efforts, these reports also serve to designate a material as a candidate for a National Disposal Site, if the material meets criteria based on quantity, degree of hazard, and difficulty of disposal. Those materials which are hazardous but not designated as candidates for National Disposal Sites, are then designated as candidates for the industrial or municipal disposal sites.

A. W. Breidenbach, Ph.D., Director
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GLOSSARY

Rad - Radiation Absorbed Dose - the absorbed dose of any nuclear radiation which is accompanied by the liberation of 100 ergs of energy per gram of absorbing material.

Rem - Roentgen Equivalent Man - a criterion of biological injury which is defined as:

Dose in rems = dose in rads x (Relative Biological Effectiveness)

$$= \text{dose in rads x } \frac{\text{physical dose of 200 -ky X-rays to produce effect of interest}}{\text{physical dose of comparison radiation to produce same effect}}$$

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PROFILE REPORT

Carbon-14, Cobalt-60, Iridium-192, Radium-226

1. GENERAL

Introduction

These four radionuclides are representative of the radioisotopes of commercial interest which are generally produced, distributed, and used by the private sector of the economy. With the exception of radium-226, these radioactive materials are either directly, or indirectly, subject to federal regulations. In many states individuals can purchase radium without proof of competence to handle it safely. It is interesting to note that two-thirds of the companies selling radium devices are located in non-agreement states and account for 91 percent of the total devices sold.[2296]

Radium-226 is a naturally occurring radionuclide first discovered in 1898. It has the longest history of use of any radioactive material and is also one of the most hazardous radionuclides. The ingestion of the luminous dial paint prepared from radium-226 was the cause of death of many of the early dial painters before the hazard was fully understood. Much of what is known of the biological effects of ionizing radiation on man is based on the effects of radium ingested by these early watch dial painters. It is estimated that over 3 million timepieces containing radium are sold annually and the exposure to the total population is probably greater than that from all other consumer products containing radioactive material. A comparison of the estimated annual radiation dose received by the critical organs from a wrist watch containing 0.15 microcuries of radium-226 with the International Commission on Radiological Protection (ICRP)[0563] annual limit for that body part is attached (Table 1).

TABLE 1

ESTIMATED ANNUAL RADIATION DOSE FROM A WRIST
WATCH CONTAINING 0.15 MICROCURIES OF RADIUM-226

Organ	Estimated Annual Dose millirem	ICRP Annual Limit For Body Part millirem 0563
Skin of the wrist	4,800	7,500
Lens of eye	110	500
Blood-forming tissue	30	500
Gonads	10	500

Radium-226 has a half-life of 1,602 years and decays by the emission of high energy, 5.68 Mev alpha particles. Radium-222 eventually decays to lead and its decay chain is attached (Table 2).

Carbon-14, cobalt-60, and iridium-192 are produced in nuclear reactors by the bombardment of a target material with neutrons. Their principal use is in the fields of nuclear medicine, radiation detection and control equipment, and in nuclear power sources. Carbon-14 has a half-life of 5,730 years and emits only a 0.156 Mev beta particle. Cobalt-60 has a half-life of 5.3 years and emits both beta and gamma particles. It decays by the emission of a 0.319 Mev beta particle to form stable nickel-60. Iridium-192 is both a beta and gamma emitter and has a half-life of 74.2 days.

Manufacture

Radium-226 is a daughter of uranium-238 and is found to occur naturally in the earth's surface. Carbon-14, cobalt-60, and iridium-192 are produced by the bombardment of a target material with neutrons. This is usually performed in a nuclear reactor. Carbon-14 is produced by the neutron bombardment of nitrogen-14. Cobalt-60 is produced from cobalt-59 and iridium-192 is produced from iridium-191.

Uses

Radium-226 has been and still is used in a wide variety of products and applications. In the past 10 years there has been a decrease in radium usage made possible by the increased availability and acceptance of use of other less hazardous radioactive materials. Radium-226 is used in producing timepieces, electron tubes, record player brushes, gauges, fire detectors, and in various self-luminous products. It is used in medicine for the treatment of tumors, superficial skin lesions, lymphoid tissue, and other diseases. It is estimated that 330 curies of radium-226 contained in 50,000 sources are used in medical applications at 2,300 facilities which provide approximately 85,000 medical treatments per year.[2296]

TABLE 2
RADIUM-226 DECAY CHAIN

Nuclide	Name	Half-Life	Major Radiation
$_{88}Ra^{226}$	Radium-226	1602 years	Alpha and gamma
$_{86}Rn^{222}$	Radon-222	3.8 days	Alpha and gamma
$_{84}Po^{218}$	Polonium-218	3.5 minutes	Alpha
$_{82}Pb^{214}$	Lead-214	26.8 minutes	Beta and gamma
$_{83}Bi^{214}$	Bismuth-214	19.7 minutes	Alpha, beta and gamma
$_{84}Po^{214}$	Polonium-214	164 miroseconds	Alpha
$_{82}Pb^{210}$	Lead-210	21 years	Beta and gamma
$_{83}Bi^{210}$	Bismuth-210	5 days	Beta
$_{84}Po^{210}$	Polonium-210	138 days	Alpha
$_{82}Pb^{206}$	Lead-206	Stable	

Carbon-14 is used in medicine to study metabolic diseases and in radioisotope gauges. Iridium-192 is widely used in the field of medicine in radiographic units. Cobalt-60 is used in isotopic power devices, in teletherapy units to treatcancer, and in radiation processing applications. Presently, there are approximately 1,830 cobalt-60 teletherapy units in use with each unit containing about 3 thousand curies of cobalt-60.[2151]

Sources and Types of Wastes

In almost all applications, these materials are used in the solid form. They are generally used as a sealed source and the material is retained within the encapsulating material. When radium-226 is used in self-luminous compounds, it is retained within the crystalline radium salt; however, there is some release of radium from luminous compounds. These materials are generally distributed throughout the country and can be found in hospitals, commercial facilities, and in households.

Physical and Chemical Properties

The physical and chemical properties of carbon-14, cobalt-60, iridium-192, and radium-226 are included in the attached worksheets. Carbon forms a vast number and variety of compounds with hydrogen, oxygen, nitrogen, and other elements. Cobalt is a brittle, hard material that resembles iron and nickel. Iridium is a metal of the platinum family. It is the most corrosion-resistant metal known and is the second heaviest known element. Radium is an alkaline metal that acts like calcium and barium chemically. It reacts with nitrogen and is mainly used in the form of salts.

2. RADIATION HAZARD

Radium-226 is one of the most hazardous radioactive materials known. Radium-226 replaces calcium in the bone structure and is a source of irradiation to the blood-forming organs. This, along with its long half-life (1,602 years) and high radiation energies, places it in the highest radiotoxicity group. It also has the longest history of use of any radioactive material, and most of the standards for the effects of ionizing radiation on man are based on this material. Carbon-14, cobalt-60, and iridium-192 are moderately dangerous radioactive materials.

The effects of their radiation exposure are primarily dependent on the amount of radiation and the portion of the body affected. The effects of whole-body gamma radiation exposure are: (1) 5 to 25 rads, minimal dose detectable by chromosome analysis or other specialized analyses, but not by hemogram; (2) 50 to 75 rads, minimal acute dose readily detectable in a specific individual (e.g., one who presents himself as a possible exposure case); (3) 75 to 125 rads, minimal acute dose likely to produce vomiting in about 10 percent of people so exposed; (4) 150 to 200 rads, acute dose likely to produce transient disability and clear hematological changes in a majority of people so exposed; (5) 300 rads, median lethal dose for single short exposure.[2666] These effects are for a single large dose of radiation or a series of substantial doses in a short interval of time to the total body. The dose delivered to a particular body organ following the inhalation of 1 microcurie of each of these radionuclides is attached (Table 3). For radium-226 the dose delivered to the bone is 300 rem following the inhalation of 1 microcuries (1.01 micrograms). The dose delivered to the bone following the injection of 1 microcurie into the body via a wound is 1,000 rem.

Standards for prolonged exposure over a 50-year period have defined the single dose limit in terms of the maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter for a radiation worker and the accumulated dose limit is 5(N - 18), where N is the individual's age in years. Limits for the thyroid, bone, and other organs have also been defined. Values of the total body burden for each radionuclide required to produce the maximum permissible dose rates defined above have been compiled.[0563] For radium-226 and carbon-14 the critical organ is the bone and the maximum permissible body burden is 0.1 and 300 microcuries, respectively. For cobalt-60 the critical organ is the total body and the maximum body burden is 10 microcuries. For iridium-192

TABLE 3

CARBON-14, COBALT-60, IRIDIUM-192, AND RADIUM-226 DOSE TO A PARTICULAR BODY ORGAN FOLLOWING INHALATION OF ONE MICROCURIE OF THE NUCLIDE

Isotope	Form	Organ	Dose rem
Carbon-14	Insoluble	Lung	0.06
Carbon-14	Soluble	Bone	0.002
Cobalt-60	Insoluble	Lung	0.77
Cobalt-60	Soluble	Total Body	0.008
Iridium-192	Insoluble	Lung	0.27
Iridium-192	Soluble	Kidney	0.05
Radium-226	Insoluble	Lung	130
Radium-226	Soluble	Bone	300

the critical organ is the kidney and the maximum body burden is 6 microcuries.

3. OTHER HAZARDS

As an element carbon is not very toxic. In the form of dust it can cause irritation of the eyes and mucous membranes. The toxicity of cobalt is also low. It can produce dermatitis and is slightly irritating to the skin. The toxicity of iridium is unknown even though soluble iridium compounds are said to be toxic.[0766] Radium's main hazard is its radioactivity. The fire and explosive hazard of the above materials is moderate.

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

Since these radionuclides are hazardous to man by inhalation, ingestion, or direct radiation exposure, great care should be exercised in their handling. Handling should be set up to prevent excessive exposure to personnel. Special procedures and adequate radiation shielding are required in their handling. Radium-226 is an alpha emitter. These alpha particles have little penetrating power and can be stopped by a sheet of writing paper. The beta particles emitted by carbon-14, cobalt-60, and iridium-192 range in energy from 0.16 to 0.67 Mev and cannot penetrate more than 0.015 to 0.085 inch of water. The gamma radiation emitted by cobalt, iridium-192, and radium-226 is highly penetrating and high-density shields, such as lead, are required to stop the radiation. For cobalt-60 which emits high energy gamma rays (1.33 Mev), approximately 3.5 inches of lead or 20 inches of concrete are required to reduce its radiation by a factor of 100. To detect and control personnel exposure to their radiation, all persons working with this material should wear dosimetry devices which directly indicate the dose. Other commonly used devices are the film badge and the thermoluminescent dosimeters (TLD).

Specially constructed containers in controlled areas should be used for storing large quantities of these materials. They should be protected by both a primary and a secondary containment barrier. Special monitoring systems and proper warning signs should be located in the general area of the storage facility. Special precautions are required in the storage of radium-226 since approximately one gram of radium produces about 0.001 milliliter of radon gas per day. Thus, stored radium should be vented to prevent the build-up of radon gas.

Radium-226 is classified as a transport group I radionuclide, cobalt-60 and iridium-192 are classified as transport group III radionuclides, and carbon-14 as a transport group IV radionuclide by the Department of Transportation. The rules and regulations governing their transportation are given in the Code of Federal Regulations (CFR) Title 14--Transportation, Parts 170 to 190.[2150] The radium-226 content is limited to 0.001 curies for a Type A package and 20 curies for a Type B package defined in 40CFR173. For cobalt-60 and iridium-192 these limits are increased to 3 and 200 curies. For carbon-14 the limits are 20 and 200 curies. The release rate of these materials is limited to zero under the specified accident conditions for Type A and B packages.[2150]

Disposal/Reuse

The disposal of these materials is governed by the AEC Manual Chapter 0524 [0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The standards for the safe release of these materials to the environment in an unrestricted area are contained in 10CFR20[2149] and their release should not result in concentrations in air and water greater than those listed in this report (Table 4). These concentrations apply to an individual and their release may be limited if a suitable sample of the population is exposed to one-third concentrations in air or water specified in this report (Table 4).

TABLE 4

CARBON-14, COBALT-60, IRIDIUM-192, AND RADIUM-226

MAXIMUM PERMISSIBLE CONCENTRATIONS[2149]

Radionuclide	Form	Concentration in Air (microcuries/milliliter)	Concentration in Water (microcuries/milliliter)
Carbon-14	Soluble	1×10^{-7}	8×10^{-4}
Carbon-14	Submersion*	1×10^{-6}	--
Cobalt-60	Soluble	1×10^{-8}	5×10^{-5}
Cobalt -60	Insoluble	3×10^{-10}	3×10^{-5}
Iridium-192	Soluble	4×10^{-9}	4×10^{-5}
Iridium-192	Insoluble	9×10^{-10}	4×10^{-5}
Radium-226	Soluble	3×10^{-12}	3×10^{-8}
Radium-226	Insoluble	2×10^{-12}	3×10^{-5}

---

*Submersion means that values given are for submersion in an infinite cloud of gaseous material.

Although rarely practiced, the disposal by release in a sanitary sewage system is limited to 0.1 microcuries of radium-226, 10 microcuries of cobalt-60, 100 microcuries of iridium-192, and 1,000 microcuries of carbon-14. The disposal by burial at any one location and time is limited to 100 times the above amounts.

The above regulations for the transportation, release, and disposal of these materials apply only to licensed materials. Radium and other accelerator-produced radionuclides are exempt from AEC control and their regulation and control is a state function. In some states the possession and use of these materials requires licensing and in others no license is required. Thus, the disposal, transportation, and use of these materials in some uses is not subject to the above regulations.

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Option No.1-Land Burial. Land burial of carbon-14, cobalt-60, and iridium-192 wastes, in small concentrations, at approved sites that are acceptable from a geologic standpoint, is an acceptable means of disposal. Their concentration should not be in excess of 10^4 times the maximum permissible concentration for the general population in 10CFR20.[2149] All wastes to be disposed of should be in a solid form and encapsulated in a suitable container. The burial site design, geology, and hydrology should be in conformance with the criteria used in selecting and licensing the present commercial burial sites.[1423] This method of disposal is not considered satisfactory for the disposal of radium-226 because radium-226 has such an extremely long half-life and a high radiotoxicity.

Option No.2-Near-Surface Solid Storage. The storage of solidified radium-226 wastes in engineered storage facilities offers the best intermediate method for storage of these wastes. The technology for these facilities has been developed and the wastes will be under surveillance and control and can be retrieved, should this be required. The wastes will be stored in stainless-steel lined concrete vaults.

Option No.3-Salt Deposits. This method offers the best potential for the disposal of radium-226 wastes since bedded salt deposits are completely free of circulating ground waters. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November of 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive waste is satisfactory.[0733] Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as an ultimate method of disposal. The critical problem is the selection of a site that meets the necessary design and geological criteria.

To summarize, small concentrations of carbon-14, cobalt-60, and iridium-192 can be disposed of by land burial at approved sites. Large concentrations of these materials, and radium-226 should be disposed of in salt deposits following storage if necessary in near-surface engineered facilities.

6. APPLICABILITY TO NATIONAL DISPOSAL SITE

Carbon-14, cobalt-60, iridium-192, and radium-226 are candidates for a National Disposal Site due to their large commercial usage and high health hazard. The recommended process for the disposal of carbon-14, cobalt-60, and iridium-192 wastes, in small concentrations, is by land burial. The recommended process for the disposal of radium-226 and large concentrations of the above radionuclides is disposal in salt beds.

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HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Carbon-14

Half-life 5730 Years

Type of Decay Negative Beta

Structural Formula $_6C^{14}$

Molecular Wt. 14   Melting Pt. 3550C   Boiling Pt. 4827C

Density 3.5 gm/cc   Specific Power

Specific Activity 4.46 curies/gm

Solubility

Cold Water insoluble   Hot Water insoluble

Others: insoluble in acid and alkali

Decay Chain

$$_6C^{14} \xrightarrow{B-} {}_7N^{14}$$

(5730 years)   (Stable)

Radiation Energy Level & Intensities

Beta: 0.155 Mev (100%)

Gamma: None

Shipping Regulations Classified as a transport group IV radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Cobalt-60

Half-life 5.3 Years

Type of Decay Negative Beta

Structural Formula

$_{27}Co^{60}$

Molecular Wt. 60 Melting Pt. 1495C Boiling Pt. 2900C

Density 8.9 gm/cc Specific Power 17.4 watts/gm

Specific Activity 1,130 curies/gm

Solubility

Cold Water insoluble Hot Water insoluble

Others: soluble in acid

Decay Chain

$$_{27}Co^{60} \xrightarrow{B-} {}_{28}Ni^{60}$$

(5.3 years) (Stable)

Radiation Energy Level & Intensities

Beta: 0.319 Mev (100%)
Gamma: 1.332 Mev (100%)
1.173 Mev (100%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation.

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Iridium-192

Half-life 74.2 days

Type of Decay Negative Beta

Molecular Wt. 192

Structural Formula

$_{77}\mathrm{Ir}^{192}$

Melting Pt. 2443C Boiling Pt. 4527C

Density 22.4 gm/cc

Specific Power

Specific Activity 9.17×10^3 curies/gm

Solubility

Cold Water insoluble Hot Water insoluble

Others: insoluble in acid and alkali, slightly soluble in aqua regia

Decay Chain

$$_{77}\mathrm{Ir}^{192} \xrightarrow{B-} {}_{76}\mathrm{Os}^{192}$$

(74d) (Stable)

Radiation Energy Level & Intensities

Beta: 0.240 Mev (8%)
0.536 Mev (41%)
0.672 Mev (46%)

Gamma: 0.296 Mev (29%)
0.308 Mev (30%)
0.317 Mev (81%)
0.468 Mev (49%)
0.589 Mev (9%)
0.604 Mev (9%)
0.612 Mev (6%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Radium-226

Half-life 1602 Years

Type of Decay Alpha

Molecular Wt. 226

Density 5 gm/cc

Structural Formula $\boxed{{}_{88}\mathrm{Ra}^{226}}$

Melting Pt. 700C

Boiling Pt. 1737C

Specific Power 1.3×10^{-4} watts/gm

Specific Activity 0.99 curies/gm

Solubility

Cold Water reacts and evolves H_2O Hot Water reacts and evolves H_2O

Others: reacts with acid

Decay Chain

$${}_{88}\mathrm{Ra}^{226} \xrightarrow{\alpha} {}_{86}\mathrm{Rn}^{222}$$

(1602 years) (3.8 days)

Decays
to Lead

Radiation Energy Level & Intensities

Alpha: 5.684 Mev (94%)
5.447 Mev (6%)

Gamma: 0.260 Mev (.007%)
0.186 Mev (4%)

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2150

PROFILE REPORT

Cesium-134, Cesium-137 (Barium-137m)

1. GENERAL

Introduction

Cesium-134 and cesium-137 are radioactive isotopes produced by the fission of uranium and plutonium and will exist in combination with other radioisotopes in high-level and low-level radioactive waste streams. Cesium-134 has a fission yield of 8 percent and cesium-137 a fission yield of 6 percent. Cesium-134 is transformed by beta decay into barium-134 which is a stable element. Cesium-137 has a half-life of 30 years and emits only beta particles. Cesium-137 is in radiation equilbrium with its short half-lived daughter (2.6 minutes), barium-137m. Barium-137m, in turn, is transformed by isometric transition (decay from an excited metastable state to lower state) into barium-317 which is a stable element.

From the viewpoint of waste management, cesium-137, because of its long half-life and high fission yield, is one of the most important fission products produced in nuclear reactors. The only fission product of comparable importance is strontium-90. The importance of cesium-137 is realized by the fact that it accounts for 10 percent of the total fission product activity in nuclear reactor wastes after 1 year, and for 52 percent of the activity after 10 years. Cesium-134 is also of significance due to its high fission yield (8 percent) and high specific power (20 watts/gram).

The projected growth in the production of cesium-134 and cesium-137 will parallel that of the civilian nuclear power program. Present projections indicate that nuclear power will account for 30 percent of the total power production by the year 1980.[2148] Annual production figures from the civilian nuclear power programs for these two isotoped have been projected to the year 2020 for light-water and fast-breeder reactor fuels (Tables 1 and 2). The expected total accumulated radioactivity of cesium-137 from the year 1970 to 2020 in millions of curies is[0705]:

Year	Radioactivity (megacuries)
1970	5
1980	1,280
1990	6,540
2000	15,600
2020	57,500

Annual production figures for the nuclear weapons program were not available.

Manufacture

Cesium-137 is one of many fission products produced directly by the fission of uranium or plutonium. The fission reaction results from the neutron bombardment of the nucleus. A typical fission reaction is:

$$_{92}U^{235} + {}_{o}n^{1} \rightarrow {}_{55}Cs^{137} + {}_{37}Rb^{96} + 2\,{}_{o}n^{1}$$

Cesium-134 is also produced in the fission reaction. Civilian nuclear power plants are the major producers and a smaller amount is produced at AEC facilities. At the present time there are 22 nuclear power plants in operation with an additional 104 being built or planned.[2151] These isotopes will also be produced in fast-breeder reactors by the fission of plutonium.

TABLE 1

CESIUM-134 CONTENT IN HIGH-LEVEL WASTES PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year	$.090\times10^5$	5.12×10^5	9.22×10^5	10.4×10^5	8.27×10^5	24.3×10^5
curies/year	$.12\times10^8$	6.66×10^8	12.0×10^8	13.6×10^8	10.8×10^8	31.6×10^8
watts/year	$.123\times10^6$	$.86\times10^6$	1.55×10^6	1.75×10^6	1.39×10^6	4.07×10^6

Fast-Breeder Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year			$.079\times10^5$	$.48\times10^5$	2.04×10^5	6.13×10^5
curies/year			$.10\times10^8$	$.62\times10^8$	2.66×10^8	8.0×10^8
watts/year			$.11\times10^6$	$.65\times10^6$	2.79×10^6	8.39×10^6

TABLE 2

CESIUM-137 CONTENT IN HIGH-LEVEL WASTES PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year	$.064x10^{6}$	$3.64x10^{6}$	$6.56x10^{6}$	$7.40x10^{6}$	$5.86x10^{6}$	$17.3x10^{6}$
curies/year	$.055x10^{8}$	$3.16x10^{8}$	$5.70x10^{8}$	$6.45x10^{8}$	$5.11x10^{8}$	$15.0x10^{8}$
watts/year	$.090x10^{5}$	$5.11x10^{5}$	$9.21x10^{5}$	$10.5x10^{5}$	$8.26x10^{5}$	$23.3x10^{5}$

Fast-Breeder Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year			$.45x10^{6}$	$2.70x10^{6}$	$11.5x10^{6}$	$34.5x10^{6}$
curies/year			$.39x10^{8}$	$2.34x10^{8}$	$10x10^{8}$	$30x10^{8}$
watts/year			$.64x10^{5}$	$3.8x10^{5}$	$16.3x10^{5}$	$49.0x10^{5}$

TABLE 3

CESIUM-134 CONTENT IN HIGH-LEVEL FUEL REPROCESSING WASTES[0705]

Light-Water Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Fission Products	After 1 year			After 10 years		
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
Cesium-134	134	1,830	175,200	6	87	8,300
Other Fission Products	34,966	8,170	2,045,000	35,096	943	308,700

Fast-Breeder Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Fission Products	After 1 year			After 10 years		
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
Cesium-134	116	220	21,200	1	11	1,000
Other Fission Products	34,984	12,780	3,408,800	34,899	755	280,000

TABLE 4

CESIUM-137 CONTENT IN HIGH-LEVEL FUEL REPROCESSING WASTES[0705]

Light-Water Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 58 MW/MTU)

Fission Products		After 1 year			After 10 years	
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
Cs^{137} - Ba^{137m}	1,200	555	203,000	980	451	165,000
Other Fission Products	32,900	9,445	2,091,000	34,120	579	152,000

Fast Breeder Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Fission Products		After 1 year			After 10 years	
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
Cs^{137} - Ba^{137m}	1,230	565	207,000	1,000	458	168,000
Other Fission Products	33,770	13,235	3,123,000	33,900	308	113,000

Physical and Chemical Properties

The physical and chemical properties of cesium-134 and cesium-137 are included in the attached worksheets. Cesium is a member of the highly electropositive alkali metal group. In its compounds, cesium has an oxidation state of +1 and is extremely reactive chemically. It is a relatively volatile metal, melting at slightly above room temperature (29.5 C) and boiling at 690 C. Most of the salts of cesium are insoluble.

2. RADIATION HAZARD

Cesium-134 and cesium-137 are moderately dangerous radioactive materials. The effects of their radiation are primarily dependent on the amount of radiation and the portion of the body affected. The effects of whole body or gamma radiation exposure are [2666]: (1) 5 to 25 rads, minimum dose detectable by chromosome analysis or other specialized analyses, but not by hemogram; (2) 50 to 75 rads, minimum acute dose readily detectable in a specific individual (e.g., one who presents himself as a possible exposure case); (3) 75 to 125 rads, minimum acute dose likely to produce vomiting in about 10 percent of people so exposed; (4) 150 to 200 rads, acute dose likely to produce transient disability and clear hematological changes in a majority of people so exposed; (5) 300 rads, median lethal dose for single short exposure. Standards for prolonged exposure over a fifty-year period have defined the single dose limit in terms of the maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter for a radiation worker and the accumulated dose limit is 5(N - 18), where N is the individual's age in years. Limits for the thyroid, bone, and other organs have also been defined.[0563]

Following the inhalation of 1 microcurie, the cesium-134 dose in the insoluble form to the lung is 0.56 rem. In the soluble form the dose delivered to the liver following the inhalation of 1 microcurie of cesium-134 is 0.09 rem. The dose delivered to the liver following injection of 1 microcurie into the body via a wound is 0.13 rem. The cesium-137 dose to the lung following the inhalation of 1 microcurie in the insoluble form is 0.46 rem, and in the soluble form the dose to the liver is 0.08 rem. The dose to the liver following the injection of 1 microcurie into the body via a wound is 0.11 rem.

Values of the maximum permissible total body burden of cesium-134 and cesium-137 which are deposited in the total body and produce the maximum permissible dose rate to a particular body organ have been compiled.[0563] For both isotopes the critical organ is the total body and the maximum permissible body burden is 20 microcuries for cesium-134 and 30 microcuries for cesium-137. The above body burden rates can be expressed in terms of the maximum permissible concentration of cesium-134 and cesium-137 in air and water at the continuous exposure rate of 168 hours per week for a period of 50 years. For cesium-134, the maximum permissible concentration in water is 9×10^{-5} microcuries per milliliter and in air 1×10^{-8} microcuries per milliliter. For cesium-137, the maximum permissible concentration in water is 2×10^{-4} microcuries per milliliter and in air 2×10^{-8} microcuries per milliliter.

3. OTHER HAZARDS

Besides its radiation hazard, cesium-134 and cesium-137 are only slightly toxic to the human body if absorbed by inhalation, ingestion, or through the skin.[0766] Their fire hazard is high, since they react with oxidizing materials and can ignite spontaneously in moist air. Their explosive hazard is moderate, but they do react with moisture to liberate hydrogen. Cesium compounds have the same toxicity as cesium unless they contain a more toxic radical.

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

Cesium-134 and cesium-137 are in the greatest abundance in the mixture of fission products which are separated from the spent fuel during processing. Since both cesium-134 and cesium-137 are hazardous to man by inhalation, ingestion, or direct radiation exposure, care

Uses

Cesium-137 is used as a radiation source for both industrial and medical equipment. Cesium-137 is used for radioisotope gauges and in industrial radiographic units. It is also used in 7 percent of the worlds total of medical teletherapy units. At present, cesium-137 is obtained from high-level waste streams in the Richland, Washington, chemical separations plant and is distributed through the Oak Ridge National Laboratory, Isotopes Sales Department. In the future, if its production is warranted by market demands, it could be available in large quantities from commercial firms operating chemical reprocessing plants. The AEC revenue from the distribution of cesium-137 through the fiscal years 1968 to 1971 is: [2151]

Year	Revenue (Dollars)
1968	83,000
1969	42,000
1970	24,000
1971	59,000

These figures illustrate the relatively small commercial market for cesium-137 at present. Cesium-134 has very limited commercial use.

Sources and Types of Wastes

Although produced in nuclear power reactors, the primary source of cesium-134 and cesium-137 is in the high-level, aqueous waste streams generated at the spent fuel processing plants. The range of chemical compositions of the various types of waste streams obtained from the processing step have been tabulated, [0715] and in all cases are aqueous solutions of inorganic nitrate salts. The characteristics of cesium-134 and cesium-137 products in the high-level wastes at two different time periods have been determined (Tables 3 and 4). These radionuclides are also found in the secondary waste streams generated at spent fuel processing plants. The activity in these wastes is quite low (less than a tenth of a curie per gal.).

is exercised in their handling. Special procedures and radiation shielding are utilized in their handling. When separated from the fission product mixture, both isotopes emit low energy beta particles which cannot penetrate more than 0.09 in. of water or 0.007 in. of lead. To stop their highly penetrating gamma rays high-density shields, such as lead, are required. To reduce their gamma radiation by a factor of 10, approximately 1.5 to 2 in. of lead or 8 to 9 in. of concrete is required. To detect and control personnel exposure, all persons working with these materials should wear dosimetry devices which directly indicate the dose. Commonly used devices are the film badge and the thermoluminescent dosimeters (TLD).

Cesium-134 and cesium-137, when separated from the other constituents of the high-level waste stream, are stored in controlled areas in specially constructed containers. They are protected by both a primary and a secondary containment barrier. Special monitoring systems and proper warning signs are located in the general area of the storage facility.

Both cesium-134 and cesium-137 are classified as a transport group III radionuclide by the Department of Transportation, and the rules and regulations governing their transportation are given in the Code of Federal Regulations (CFR) Title 49--Transportation, Parts 170 to 190.[2150] Their content is limited to 3 curies for a Type A package and 200 curies for a Type B package defined in 49CFR173. The limits are increased to 20 to 5,000 curies if their physical form meets the requirements of a special form material. Their release rate is limited to zero under the specified accident conditions for Type A and B quantities.[2150] The allowable release of radioactivity from packages containing large quantities of these isotopes is limited to gases and contaminated coolant containing total radioactivity exceeding neither 0.1 percent of the total radioactivity of the package nor 10 curies under the hypothetical accident conditions prescribed in 49CFR173.

The bulk of the cesium-134 and cesium-137 arrives at the spent fuel processing plants in the spent fuel assemblies which are contained in Type B packages. The cesium-134 and cesium-137 may be stored as a liquid at the processing plant for up to 5 years and are then solidified and leave the plant in Type B packages.

Disposal/Reuse

After solidification at the fuel processing facility, the bulk of the cesium wastes may be held for up to 5 years before being shipped to a Federal repository. This repository has not yet been defined but work is in progress, under AEC sponsorship, on engineered surface storage facilities which will be designed to store solidified high-level waste for up to 100 years. In the interim, a satisfactory ultimate disposal scheme, such as disposal in salt deposits, will be evolved.

The disposal of cesium-134 and cesium-137 is governed by the AEC Manual Chapter 0524[0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The radiation protection standards for these two groups are attached (Table 5). The standards for the safe disposal of cesium-134 and cesium-137 also define their maximum concentrations in air and water (Table 6).

The disposal of cesium-134 by release into a sanitary sewage system is limited to 10 microcuries and the disposal of cesium-137 is limited to 100 microcuries. The disposal by burial in the soil at any one location and time is limited to 1,000 microcuries of cesium-134 and 10,000 microcuries of cesium-137.[2149]

TABLE 5

RADIATION PROTECTION STANDARDS FOR INDIVIDUALS AND POPULATION GROUPS FOR EXTERNAL AND INTERNAL EXPOSURE[2149]

Type of Exposure	Dose to Individuals at Points of Maximum Probably Exposure (rem per year)	Average Dose to a Suitable Population Sample (rem per year)
Whole body, gonads, or bone marrow	0.5	0.17
Thyroid or bone	1.5	0.5
Bone (alternate standards)	Body burden at 0.003 micrograms of radium 226 or its biological equivalent	Body burden of 0.001 micrograms of radium 226 or its biological equivalent

TABLE 6

CESIUM-134 AND CESIUM-137 MAXIMUM PERMISSIBLE CONCENTRATIONS[2149]

Isotope	Exposure Group	Form	Concentration in Air (microcuries/milliliter)	Concentration in Water (microcuries/milliliter)
Cs-134	Individual	Soluble	1×10^{-9}	9×10^{-6}
Cs-134	Individual	Insoluble	4×10^{-10}	4×10^{-5}
Cs-134	Population	Soluble	$.33 \times 10^{-9}$	3×10^{-6}
Cs-134	Population	Insoluble	1.3×10^{-10}	1.3×10^{-5}
Cs-137	Individual	Soluble	2×10^{-9}	2×10^{-5}
Cs-137	Individual	Insoluble	5×10^{-10}	4×10^{-5}
Cs-137	Population	Soluble	$.67 \times 10^{-9}$	$.67 \times 10^{-5}$
Cs-137	Population	Insoluble	1.7×10^{-10}	1.3×10^{-5}

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Recovery

Small quantities of cesium-134 and cesium-137 may be recovered from the high-level waste streams for industrial purposes. Another potential advantage to recovery is that two or more radionuclides with similar characteristics could undergo the same handling, storage, or disposal processes. Although presently rarely recovered, various techniques, in various stages of development, are being investigated.

Option No. 1 - Precipitation. Cesium-134 and cesium-137 can be removed from the high-level reactor wastes by precipitating with nickel ferrocyanide. Over 99 percent of the cesium can be removed satisfactorily up to a pH of 10.[0553,0714] Cesium can also be removed by precipitating with phosphotungstate[0714] or absorbing on alumino-silicate zeolites.[0738] Cesium can be further purified by ion exchange. Cesium is recovered from the other high-level wastes for commercial purposes or for safety reasons.

Option No. 2 - Scavenging-Precipitation Foam Separation. The removal of cesium-134 and cesium-137 from low-level radioactive waste water by scavenging-precipitation foam separation has been studied by Oak Ridge National Laboratory.[0703] The process consists of two steps: (1) precipitating in a sludge-blanket clarification step; and (2) achieving final decontamination in a foam separation column. The precipitation step includes the use of Grundite clay for the sorption of cesium. The Grundite clay increased the cesium decontamination factor (ratio of initial to final concentration) approximately nine times. The overall decontamination factor for cesium was eight. At present, further development is required to reduce operating costs and increase processing rates.

Option No. 3 - Scavenging-Precipitation Ion Exchange. In this process final decontamination is obtained by ion exchange columns from the scavenging-precipitation process. The process includes a provision for the recycle of the ion exchange waste to the scavenging-precipitation step. All the removed radionuclides are concentrated in the clarifier sludge. With most ion exchange resins sodium is removed with cesium. Cesium can be separated from sodium by phenolic-base cation exchanges at high pH values. For this process the overall decontamination factor for cesium varied from 100 to 3,000.[0705] The use of ion exchange resins with a scavenging-precipitation step is a fairly simple and efficient means of removing cesium-134 and cesium-137 from low-level radioactive aqueous wastes.

Option No. 4 - Water Recycle. The water recycle process is used for decontaminating radioactive waste water and recycling the purified water for reuse. This process has been demonstrated at the pilot plant scale by Oak Ridge National Laboratory.[0707] The steps in the process include: (1) clarification by the addition of coagulants; (2) demineralization by cation-anion exchange; and (3) sorption on granular activated carbon. The majority of the cesium is removed by the cation-anion exchange. The cesium overall decontamination factor was 14,000. The method is an improvement to the treat-and-discharge methods, but further work is required until full-scale production use is obtained.

Storage/Disposal

Option No. 1 - Land Burial. Land burial of cesium-134 and cesium-134 in low-level wastes, at approved sites that are acceptable from a geologic and hydrologic standpoint, is an acceptable means of disposal. The cesium-134 and cesium-137 concentrations should not be in excess of 10^4 times their maximum permissible concentrations for the general population in 10CFR20.[2149] All cesium wastes to be disposed of should be in a solid form and encapsulated in a suitable container. Liquid wastes should be solidified, preferably using asphalt, in accordance with the methods described in the Radioactive Waste Solidification report. The burial trenches should be designed not to intercept the ground water table and constructed with a bottom drain and sump for water monitoring. The trenches should be covered with either asphalt or vegetation to limit infiltration of water. The burial site design, geology, and hydrology should be in conformance with the criteria used in selecting and licensing the present commericial burial sites.[1423] Since land burial sites meeting the criteria are operating on a commercial scale currently, they are considered the most satisfactory method of disposing of dilute concentrations.

Option No.2 - Near-Surface Liquid Storage. Near-surface storage (using carbon steel and stainless steel tanks encased in concrete and buried underground) of high-level, reactor-produced aqueous solutions of cesium and other fission product salts is not considered as a satisfactory means of storage. Aqueous solutions of high-level wastes from reactor fuelds have been stored in this manner over the past 25 years. The tanks range in size from 0.33 to 1.3 million gal. and are equipped with devices for measuring temperatures, liquid levels, and leaks. These tanks are considered as an interim storage technique due to a general lack of confidence in their long-term integrity.[0705] Therefore, the near-surface storage of aqueous solutions of cesium and other fission product salts in steel tanks should only be considered as a near-term storage technique and not as a permanent storage or disposal technique.

Option No.3 - Near-Surface Solid Storage. The storage of high-level solidified cesium-134 and cesium-137 wastes and other waste salts in engineered storage facilities offers the best method for intermediate storage of these concentrated wastes. The advantages of this method are that the wastes will be under surveillance and control and that they can be retrieved, should this be required. The high-level wastes from fuel processing should be solidified and packaged in a suitable container (steel). Of the four high-level solidification processes developed, spray or phosphate glass solidification processes offer better solidified waste characteristics than the pot calcination or fluidized bed calcination processes (see Radioactive Waste Solidification report). The wastes will be stored in stainless steel-vaults encased in concrete and will be either air or water cooled.

Option No. 4 - Salt Deposits. This method offers the greatest potential for the disposal of high-level wastes since bedded salt deposits are completely free of circulating ground waters. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November of 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive wastes is satisfactory.[0733] Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as a method of ultimate disposal. The cesium wastes must be solidified and disposed of in the solid form, encapsulated in a suitable container. The solidified wastes are buried in rooms carved in the salt deposits approximately 1,000 ft below the ground. The salt is a good heat transmitter, provides about the same radioactive shielding as concrete, and can heal its own fractures by plastic flow. The critical problem is the selection of a site that meets the necessary design and geological criteria.

Option No. 5 - Bedrock Disposal. The disposal of high-level, aqueous solutions of cesium-134 and cesium-137 wastes, along with other radioactive wastes in vaults excavated in crystalline rock over 1,500 ft beneath the ground is currently being evaluated by E. I. du Pont de Nemours, at their Savannah River Plant near Aiken, South Carolina.[0894,1396] The wastes would be stored in six tunnels and once in the tunnels the wastes will seep into the surrounding rock. Located above the crystalline rock is the Tuscaloosa formation, a good source of fresh water, which is separated by a layer of clay that would act as a barrier to the leakage of radioactive wastes. An advisory committee appointed by the National Academy of Sciences recommended abandonment of the project. In May 1972, another National Academy of Sciences panel concluded that bedrock storage provides a reasonable prospect for long-term safe storage, but precise information is needed to decide if and where underground storage vaults should be built. Another method has also been proposed for disposing of liquid wastes by in situ incorporation in molten silicate rock.[2146] At the present time both of these methods are unproven since sufficient engineering data or exploration has not been completed to verify their suitability. Due to the long half-lives of certain of the elements in the mixed fission products and actinides, it is doubtful that data could ever be accumulated to prove that the geological characteristics of the site over the next few hundred years are acceptable to ensure the absolute safety of such a disposal method.

Option No. 6 - Hydraulic Fracturing. The direct disposal of aqueous, low-level radioactive wastes into shale formations has been investigated by Oak Ridge National Laboratory.[0709] The method consists of mixing the aqueous wastes with cement and pumping the resulting slurry down a well out into a nearly horizontal fracture in a thick shale formation. Additional work is required to demonstrate that this method of disposal is satisfactory.

To summarize, cesium-134 and cesium-137 can be recovered from the high-level waste streams for separate disposal or reuse. The acceptable method of treatment is solidification, followed by storage in a near-surface engineered storage facility and ultimate disposal in a salt deposit. Cesium should be recovered from the low-level waste streams to minimize the amount directly released to the environment. The recovered cesium should be solidified, preferably using asphalt, and disposed of at approved sites by land burial.

6. APPLICABILITY TO NATIONAL DISPOSAL SITES

Cesium-134 and cesium-137 are candidates for a National Disposal Site due to their public health hazard and projected growth with that of the civilian nuclear power program. From a cost and safety viewpoint, it would be desirable to combine the reprocessing plant and the

National Disposal Site. Interim storage in near-surface engineered facilities offers considerable latitude in the site selection. The site selection for ultimate disposal of these wastes is limited to particular geological areas in which salt deposits are present. Temporary, or interim, storage prior to transfer to a National Disposal Site could be accomplished at many site locations.

The recommended treatment for low-level waste streams is recovery followed by solidification with asphalt and disposed by land burial.

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HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Cesium-134

Structural Formula

$_{55}Cs^{134}$

Half-life 2.05 years

Type of Decay Negative Beta

Molecular Wt. 134 Melting Pt. 28.5C Boiling Pt. 690C

Density 1.88 gm/cc Specific Power 20.4 watts/gm

Specific Activity 1,220 curies/gm

Solubility

Cold Water Reacts Hot Water Reacts

Others: Soluble in liquid NH_3

Decay Chain

$$\underset{(2.05y)}{Cs^{134}} \xrightarrow{B^-} \underset{(stable)}{Ba^{134}}$$

Radiation Energy Level & Intensities

Beta: 0.662 Mev (71%)
0.410 Mev (1%)
0.089 Mev (28%)
Gamma: 1.365 Mev (3.4%)
1.168 Mev (1.9%)
0.769 Mev (99%)
0.605 Mev (98%)
0.570 Mev (23%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Cesium-137

Half-life 30 years

Type of Decay Negative Beta

Structural Formula

${}_{55}Cs^{137}$

Molecular Wt. 137

Melting Pt. 28.5 C

Boiling Pt. 690 C

Density 1.88 gm/cc

Specific Power 0.42 watts/gm

Specific Activity 87 curies/gm

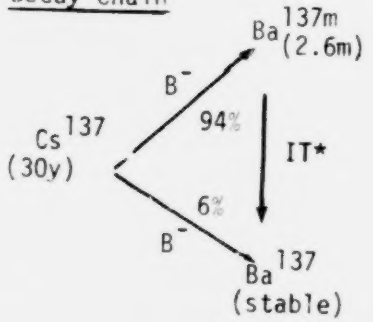
Solubility

Cold Water Reacts

Hot Water Reacts

Others: Soluble in liquid NH_3

Decay Chain



Radiation Energy Level & Intensities

Cesium-137

Beta: 1.176 Mev (6%)
0.514 Mev (94%)

Barium-137

Gamma: 0.662 Mev (39%)

*IT = isometric transition

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2147
(3) 2150

PROFILE REPORT

Hydrogen-3

1. GENERAL

Introduction

Tritium, an isotope of hydrogen, is produced in nuclear reactors in substantial quantities. It is currently released to the environment in the form of tritiated coolant water from the reactor and tritium wastes resulting from processing of the spent nuclear fuels. Although it is one of the least hazardous radioactive nuclides, the recovery and retention of tritium may be required in the future due to its long half-life for radioactive decay, the rapid rate of expansion in the nuclear power and fuel-reprocessing industries, and its ability to be metabolized in the form of tritiated water and incorporated into body fluids and tissues.

Tritium is produced in the fission of uranium-235 and plutonium-239 with yields of 0.01 percent and 0.02 percent, respectively. Tritium decays by the emission of a beta particle and an anti-neutrino to form stable helium-3. The half-life for this process is approximately 12.3 years. The average energy of the beta emitted by tritium is 5.6 Kev, which is only about one one-hundredth of the energy emitted by most other beta emitters.

The projected growth in production of this isotope will parallel that of the civilian nuclear power program. Present projections indicate that nuclear power will account for 30 percent of the total power production by the year 1980.[2148] The expected annual production and accumulated wastes of hydrogen-3 from the civilian nuclear power program are included (Table 1).

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TABLE 1

HYDROGEN-3 GENERATED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual			Calendar Year			
Production	1970	1980	1985	1990	2000	2020
grams/year	3.74	213	384	434	344	1010
curies/year	$.036 \times 10^6$	2.06×10^6	3.72×10^6	4.21×10^6	3.34×10^6	9.80×10^6
watts/year	1.29	73.4	132	149	119	348

Fast-Breeder Reactor Fuels

Annual			Calendar Year			
Production	1970	1980	1985	1990	2000	2020
grams/year			34.2	207	881	2650
curies/year			$.33 \times 10^6$	2.01×10^6	8.55×10^6	26.8×10^6
watts/year			11.8	71.1	304	914

Uses

Tritium is used in luminous devices, tagging, and fusion research.

Sources and Types of Wastes

Tritium may be produced in nuclear reactors by one or more of the following five methods: fissioning of uranium, neutron capture reactions with boron and lithium added to the reactor coolant, neutron capture reactions with boron in control rods, activation of deuterium in water, and high energy neutron capture reactions with structural materials.[2378]

Fission-product tritium is generally contained by the cladding surrounding each fuel element. While small amounts of tritium were found in the past to leak to the primary coolant with the stainless-steel clad elements, the use of zirconium cladding has been found to limit the diffusion of tritium through the cladding to very low quantities. The fission-product tritium generated is contained until the spent fuel is processed, when it appears as tritiated water in the fuel processing plant evaporator condensates. Additional sources of tritium are in the primary coolant loops of pressurized water reactors and high temperature gas-cooled reactors. In the pressurized water reactors, boric acid which is dissolved in the primary coolant for reactivity control causes the production of tritium by neutron capture reactions. The production of tritium occurs in high temperature gas cooled reactors by ternary fission and by activation of helium-3 found in trace amounts in the helium coolant. Although the primary coolant from these reactors is not routinely discharged to the waste disposal system, leakage of coolant from pumps and the water utilized during the refueling operation are discharged to the waste disposal systems. In the reactor power plant waste disposal systems the water undergoes several treatment processes before being released to the environment, but none of these processes is effective in removing tritium. Tritium may compose between 50 and almost 100 percent of the total amount of radioactive material discharged as liquid waste from nuclear reactors. The amount of tritium discharged from reactors in a gaseous form is only about 1 percent of the total tritium discharge.

Physical and Chemical Properties

The physical and chemical properties of tritium are included in the attached worksheet. It is found as a colorless gas and in the form of tritiated water. It is undetectable by conventional methods of gross radioactivity analysis. Special analytical techniques such as liquid scintillation counting must be used to measure tritium.

2. RADIATION HAZARD

Tritium is a moderately hazardous radioactive material. Because it is an isotope of hydrogen, it can be metabolized in the form of tritiated water and incorporated into body fluids and tissues.

The effects of radiation exposure are primarily dependent on the amount of radiation and the portion of the body affected. A single ingestion of tritiated water having an activity of 1 microcurie will produce a total dose to the body tissues of 0.21 rem. Continuous ingestion of water having a specific activity of 1 microcurie of tritium per milliliter will produce a dose rate of 170 rem per year to body tissues.

Standards for prolonged exposure over a 50-year period have defined the single dose limit in terms of the maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter for a radiation worker and the accumulated dose limit is 5(N-18), where N is the individual's age in years.

3. OTHER HAZARDS

Tritium has negligible toxicity other than the radiation hazard. Tritium is flammable in air and mixtures with air are explosive.

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

Since tritium is moderately hazardous to man by inhalation or ingestion, care must be exercised in its handling. Handling must be set up to prevent excessive exposure to personnel and, in fact, to prevent any unnecessary exposure.

These materials should be stored in controlled areas in specially constructed containers. Tritium, as a gas, as luminous paint, or adsorbed on solid material is classified as a group VII radionuclide by the Department of Transportation. The rules and regulations governing its transportation are given in the Code of Federal Regulations (CFR) Title 49--Transportation, Parts 170 to 190.[2150] The transport group VII radionuclide content is limited to 1,000 curies for a Type A package and 50,000 curies for a Type B package defined in 49CFR173. Their release rate is limited to zero under the specified accident conditions for Type A and B quantities.[2150]

Disposal/Reuse

The disposal of these materials is governed by the AEC Manual Chapter 0524[0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The standards for the safe release of this material to the environment in an unrestricted area are contained in 10CFR20[2149] and its release should not exceed the concentrations in air and water listed in this report (Table 2). These release rates apply to an individual and its release may be further limited if a suitable sample of the population is exposed to one-third the concentrations in air or water specified in this report (Table 2).

TABLE 2

TRITIUM ENVIRONMENT RELEASE RATES[2149]

Fission Product	Form	Concentration in Air (microcuries/milliliter)	Concentration in Water (microcuries/milliliter)
Hydrogen-3	Soluble	2×10^{-7}	3×10^{-3}
	Insoluble	2×10^{-7}	3×10^{-3}
	Submersion*	4×10^{-5}	---

*Submersion means that the values given are for submersion in a semispherical infinite cloud of airborne material.

Although rarely practiced, the disposal by release in a sanitary sewage system is limited to 10,000 microcuries.[2149] The disposal by burial at any one location and time is limited to 100 times the above amount.

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Recovery

Tritium is not currently recovered at the reactor site or the spent fuel processing plant site. However, the Oak Ridge National Laboratory is conducting experimental work on recovering tritium during fuel reprocessing for the fast breeder reactor. Voloxidation is the term for the tritium recovery process being investigated in which the fuel, after being chopped in the head-end process, is heated to drive off the tritiated hydrogen. By collecting and condensing the effluent, greater than 99 percent of the tritium is recovered. It is feasible that such a process could be incorporated into the reprocessing of fuel from the present, light water reactors.

Storage/Disposal

The current method of tritium disposal is dilution and dispersion, both at the reactor site and the spent fuel processing plant site. Most of this release is in the form of tritiated water in the liquid waste streams and only a very small percent is released to the air. Tritium discharge concentrations are usually much less than 1 percent of the discharge limits. Because of the inherently small radiological hazard from tritium, the difficulty of removing tritium from the reactor's tritiated water, and the small percentage of the allowable concentrations that are currently being released to the environment at the reactor site, tritium removal is probably not practical in the near future. At the spent fuel processing plant, however, a process similar to the voloxidation process should be incorporated in order to recover tritium and dispose of it by one of the following methods.

Option No. 1 - Land Burial. Land burial of tritium wastes, in small concentrations, at approved sites that are acceptable from a geologic and hydrologic standpoint, is an acceptable means of disposal. Its concentrations should not be in excess of 10^4 times its maximum permissible concentrations for the general population in 10CFR20.[2149] The burial trenches should be designed not to intercept the ground water table and constructed with a bottom drain and sump for water monitoring. The trenches should be covered with either asphalt or vegetation to limit infiltration of water. The burial site design, geology, and hydrology should be in conformance with the criteria used in selecting and licensing the present commercial burial sites.[1423] Since land burial has successfully been practiced on a commercial scale, it should be considered as the most satisfactory method of disposing of low concentrations of these wastes.

Option No. 2 - Near Surface Storage. The storage of high concentrations of these wastes in engineered surface facilities offers the best intermediate method for storage of these wastes. The technology for these facilities has been developed and the wastes will be under surveillance and control and can be retrieved, should this be required. The wastes will be stored in stainless-steel lined concrete vaults which will be either air or water cooled.

Option No. 3 - Salt Deposits. This method offers the best potential for the disposal of this radionuclide. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive wastes is satisfactory.[0733] Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as an ultimate method of disposal. The wastes are buried in rooms carved in the salt deposits approximately 1,000 ft below the ground. The salt is a good heat transmitter, provides about the same radioactive shielding as concrete, and can heal its own fractures by plastic flow. The critical problem is the selection of a site that meets the necessary design and geological criteria.

To summarize, the majority of the tritium generated by the nuclear power reactors is currently released to the environment in the form of liquid wastes. Most of this release occurs at the fuel reprocessing plant, where the tritium could be recovered by voloxidation or a similar process. The tritium should be encapsulated and disposed of in salt deposits following storage in engineered storage facilities.

6. APPLICABILITY TO NATIONAL DISPOSAL SITES

Tritium is a candidate for a National Disposal Site due to its projected growth with that of the civilian nuclear power program. It should be recovered and encapsulated at the fuel reprocessing plant and shipped to the National Disposal Site for disposal in salt beds following storage in engineered storage facilities.

7. REFERENCES

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0705. Staff of the Oak Ridge National Laboratory. Siting of fuel reprocessing plants and waste management facilities. ORNL-4451, 1970. 500 p.

0766. Sax, N. I. Dangerous properties of industrial materials. 3d ed., New York, Reinhold Publishing Corp., 1968. 1,251 p.

2148. Atomic Energy Commission. Forecase of growth of nuclear power, Jan. 1971. Washington, U. S. Government Printing Office, WASH-1139, 1971. 187 p.

2149. Code of Federal Regulations. Title 10--atomic energy. (Revised as of Jan. 1, 1971). Washington, U. S. Government Printing Office, 1971. 429 p.

2150. Code of Federal Regulations. Title 49--transportation, parts 1 to 199. (Revised as of Jan. 1, 1971). Washington, U. S. Government Printing Office, 1971. 952 p.

2378. Selected materials on environmental effects of producing electric power. Report of the Joint Committee on Atomic Energy Congress of the United States. 91st Cong., 1st sess., Aug. 1969. Washington, U. S. Government Printing Office. 553 p.

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Tritium

Half-life 12.26 years

Tyne of Decay Negative Beta

Molecular Wt. 6.05 Melting Pt. ______ Boiling Pt. ______

Density ______ Specific Power ______

Specific Activity 9700 curies/gm

Structural Formula

$_1H^3$

Solubility

Cold Water ______ Hot Water ______

Others: ______

Decay Chain

$$\underset{(12.26y)}{H^3} \longrightarrow \underset{(stable)}{He^3}$$

Radiation Energy Level & Intensities

Beta: 5.6 Kev (average)
18.6 Kev (maximum)

Shipping Regulations Classified as a transport group VII radionuclide by the Department of Transportation

Comments ______

References:
(1) 0766
(2) 2150
(3) 2378

PROFILE REPORT

Iodine-129, Iodine-131, Krypton-85, Xenon-133

1. GENERAL

Introduction

Iodine and the noble gases, krypton and xenon, are produced during the fission of uranium in nuclear reactors. They represent a potential source of environmental contamination since they are presently released to the environment during reprocessing of the nuclear fuels. Although the release rates at present reprocessing plants are low, the recovery and retention of these gases may be required in the near future due to the rapid rate of expansion of the nuclear power industry and the corresponding expansion in the nuclear fuel reprocessing industry. In addition to the anticipated growth in this industry, economic incentives exist for reducing fuel decay times prior to reprocessing. Present reprocessing plants handle 150-day decayed light water reactor fuel. With the introduction of the fast-breeder reactors it is economically desirable to reprocess the fuel after 30 days decay. The shortened decay period will increase the reprocessing plant off-gas handling requirements for the short-lived isotopes such as iodine-131 (8 days) and xenon-133 (5 days). The iodine-131 activity in 30-day-old reactor fuel is 36,000 times higher than it is in 150-day-old reactor fuel.

Of the fission-product halogens only the isotopes iodine-129 and iodine-131 are physiologically significant after 30 days of post-irradiation decay. The iodine-131 contents of reactor fuels are approximately 72,000 and 2 curies per metric ton of reactor fuel after decay times of 30 and 150 days respectively. The iodine-129 content is only about 0.03 curies per metric ton of reactor fuel. Even though the iodine-129 content in reactor wastes is low, it is significant since it has a half-life of

17 million years compared to the iodine-131 half-life of 8 days. Iodine-129 and iodine-131 are also reconcentrated by biological processes in the food chain leading to man. This reconcentration occurs in the grass-cow-milk pathway to the thyroids of small children and man.

Krypton and xenon are both produced in significant quantities in nuclear reactors. These isotopes are chemically inert and once released they do not concentrate in body tissues. The various isotopes of krypton and xenon in reactor fuels are attached (Table 1). The only two isotopes of significance are krypton-85 and xenon-133. Krypton-85 has a half-life of 10.8 years, a moderate beta radiation energy, a low gamma radiation level, and a fission yield of 1.3 percent. Xenon-133 has a half-life of 5.3 days, a moderate beta and gamma radiation energy, and a fission yield of 6.6 percent.

The projected growth in production of these isotopes will parallel that of the civilian nuclear power program. Present projections indicate that nuclear power will account for 30 percent of the total power production by the year 1980.[2148] The expected annual production of iodine-129 (Table 2) and the expected annual production and accumulated wastes of krypton-85 (Figure 1) from the civilian nuclear power program are included. Production figures for iodine-131 and xenon-133 are not included since due to their short half-life the amount present varies with the post-irradiation decay time.

Uses

Krypton-85 is used in the preparation of self-illuminating materials and devices. Its properties also make it useful in leak detection equipment, thickness gauges, and in gas chromatography. The fission product xenon, including xenon-133, which is essentially non-radioactive by the time it is recovered, is used in scintillation devices because it has a higher proportion of heavy isotopes of xenon than does naturally occurring xenon. Xenon is also used in the manufacture of light bulbs that have an extra long life. Iodine-131 is used in medical diagnosis and therapy and also in agricultural research. Since iodine-131 tends to

TABLE 1

THERMAL-NEUTRON FISSION YIELD OF KRYPTON AND XENON FROM U-235 REACTOR FUELS

Fission Products	Fission Yield Percent	Half-Life
Krypton-83	0.544	Stable
Krypton-84	1.00	Stable
Krypton-85	1.293	10.76 years
Krypton-86	2.02	Stable
Xenon-131	2.93	Stable
Xenon-132	4.38	Stable
Xenon-133	6.62	5.3 days
Xenon-134	8.06	Stable
Xenon-135	6.30	9.2 hours
Xenon-136	6.46	Stable

TABLE 2

IODINE-129 CONTENT IN WASTE PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year	$.012 \times 10^6$	$.69 \times 10^6$	1.24×10^6	1.4×10^6	1.11×10^6	3.26×10^6
curies/year	1.97	112	202	229	182	534
watts/year	.0085	.0485	.0874	.099	.078	.231

Fast-Breeder Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year			$.116 \times 10^6$	$.70 \times 10^6$	3.0×10^6	9.0×10^6
curies/year			19.4	117	500	1500
watts/year			.0083	.051	.215	.649

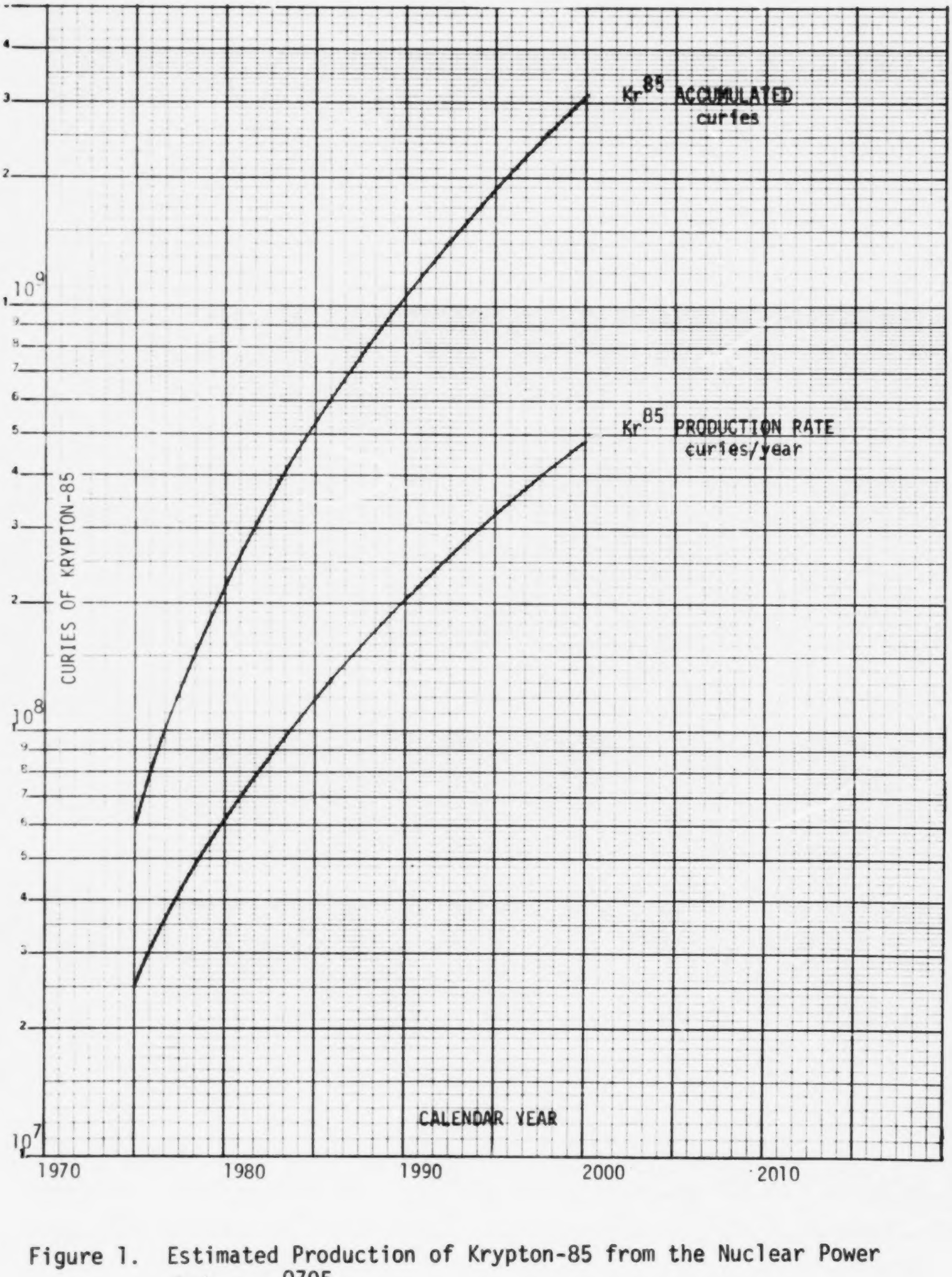

Figure 1. Estimated Production of Krypton-85 from the Nuclear Power Industry.0705

concentrate in the thyroid, it is used in the treatment of hyperthyroidism and in treating congestive heart failure. Iodine-129, due to its long half-life, has very limited commercial use.

Sources and Types of Wastes

Iodine, krypton, and xenon are produced in nuclear reactors and are contained within the spent fuel elements. The spent fuel elements are removed from the reactor and processed for the recovery of usable fissionable materials. In the spent fuel processing plants the majority of these radionuclides are released as gases during the dissolution of the fuel elements in the head-end process. They are also released in the venting of process vessels and in the treatment of liquid wastes. Iodine-129 and iodine-131 are released in the off-gas streams in the form of elemental iodine (I_2), hydrogen iodide, or as organic iodide compounds such as methyl iodide. Krypton and xenon are released in their elemental form and do not form compounds with other materials since they are chemically inert.

Physical and Chemical Properties

The physical and chemical properties of iodine, krypton, and xenon are included in the attached worksheets. Iodine is a member of the halogen group and is a solid at room temperature. Iodine is easily melted and vaporized and is a very powerful oxidizing agent. Iodine forms compounds with many elements, but is less active than the other halogens which displace it from iodide compounds. Krypton and xenon are members of the rare or inert gas group. These gases are characterized generally by their zero valence and are considered very inert. Some relatively unstable compounds of these elements are known, however.

2. RADIATION HAZARD

Iodine, xenon, and krypton are moderately hazardous radioactive nuclides. Krypton and xenon are chemically inert and once released they do not concentrate in body tissues. Iodine does reconcentrate by the grass-cow-milk pathway to the thyroid.

The effects of their radiation exposure are primarily dependent on the amount of radiation and the portion of the body affected. The effects of whole-body gamma radiation exposure are: (1) 5 to 25 rads, minimal dose detectable by chromosome analysis or other specialized analyses, but not by hemogram; (2) 50 to 75 rads, minimal acute dose readily detectable in a specific individual (e.g., one who presents himself as a possible exposure case); (3) 75 to 125 rads, minimal acute dose likely to produce vomiting in about 10 percent of people so exposed; (4) 150 to 200 rads, acute dose likely to produce transient disability and clear hematological changes in a majority of people so exposed; (5) 300 rads, median lethal dose for single short exposure.[2666] The effects are for a single large dose of radiation or a series of substantial doses in a short interval of time to the total body. The dose delivered to a particular body organ following the inhalation of 1 microcurie of each of these radionuclides is attached (Table 3). For iodine-131 a dose of 25 rem is delivered to the thyroid following the inhalation of 15.6 microcuries (0.002 micrograms).

Standards for prolonged exposure over a 50-year period have defined the single dose limit in terms of the maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter for a radiation worker and the accumulated dose limit is 5(N - 18), where N is the individual's age in years. Limits for the thyroid, bone, and other organs have also been defined. Values of the total body burden for each radionuclide required to produce the maximum permissible dose rates defined above have been compiled.[0563] For iodine the critical organ is the thyroid and the maximum permissible body burden is 2 microcuries for iodine-131 and 9 microcuries for iodine-129.

3. OTHER HAZARDS

Besides its radiation hazard iodine is moderately toxic whereas krypton and xenon are only slightly toxic. Iodine contact with the skin can cause lesions and iodine vapor is intensely irritating to the eyes and mucous membranes. The recommended maximum allowable concentration of iodine in the air is 1 mg/cu. meter.[0766]

TABLE 3

IODINE, KRYPTON, AND XENON DOSE TO A PARTICULAR BODY ORGAN FOLLOWING INHALATION OF ONE MICROCURIE OF THE NUCLIDE

Isotone	Form	Organ	Dose rem
Iodine-129	Insoluble	Lung	0.09
Iodine-129	Soluble	Thyroid	7.0
Iodine-131	Insolbule	Lung	0.03
Iodine-131	Soluble	Thyroid	1.6
Krypton-85	Insoluble	Lung	0.26
Xenon-133	Insoluble	Lung	0.02

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

Since these radionuclides are hazardous to man by inhalation, ingestion, or direct radiation exposure, care is exercised in their handling. Handling is set up to prevent excessive exposure to personnel. Special procedures and radiation shielding are utilized in their handling. The beta particles emitted by iodine, xenon, and krypton are of moderate energy and cannot penetrate more than 0.1 inches of water. Their gamma radiation is highly penetrating and high-density shields, such as lead, are required to stop the radiation. To detect and control personnel exposure to their radiation all persons working with this material should wear dosimetry devices which directly indicate the dose. Commonly used devices are the film badge and the thermoluminescent dosimeters (TLD).

These materials are stored in controlled areas in specially constructed containers. They are protected by both a primary and a secondary containment barrier. Special monitoring systems and proper warning signs should be located in the general area of the storage facility.

Iodine-129 and iodine-131 are classified as a transport group III radionuclide and krypton-85 and xenon-133 are classified as a group IV radionuclide in the uncompressed state, at a pressure less than 14.7 psia, and as a group III radionuclide at pressures greater than 14.7 psia by the Department of Transportation. The rules and regulations governing their transportation are given in the Code of Federal Regulations (CFR) Title 49--Transportation, Parts 170 to 190.[2150] The transport group III radionuclide content is limited to 3 curies for a Type A package and 200 curies for a Type B package defined in 49CFR173. Their release rate is limited to zero under the specified accident conditions for Type A and B quantities.[2150]

Disposal/Reuse

The disposal of these materials is governed by the AEC Manual Chapter 0524[0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The standards for the safe release of these materials to the environment in an unrestricted area are contained in 10CFR20[2149] and their release should not cause the concentrations in air and water to exceed the concentrations listed in this report (Table 4). These concentrations apply to an individual and their release may be further limited if a suitable sample of the population is exposed to one-third the concentrations in air or water specified in this report (Table 4).

Although rarely practiced, the disposal by release in a sanitary sewage system is limited to 1 microcurie for iodine-129, 10 microcuries for iodine-131 and 1,000 microcuries for krypton-85 and xenon-133.[2149] The disposal by burial at any one location and time is limited to 100 times the above amounts.

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Iodine Recovery

Option No.1-Caustic Scrubbers. Iodine has been removed from gas streams in many applications by scrubbing with caustic solutions and reacting with silver nitrate impregnated on ceramic packing. Caustic scrubbers are effective in removing approximately 90 percent of the iodine in the off-gas streams and the silver nitrate towers remove about 99 percent of the remaining iodine. In general, caustic scrubbers are not effective in removing organic iodides, such as methyl iodide. Organic iodides can be effectively removed by using an aqueous scrubbing medium of mercuric nitrate in nitric acid if sufficient contact time is allowed for the relatively slow reaction between the absorbed organic iodide and mercury ion.

TABLE 4

IODINE, KRYPTON, AND XENON NATIONAL PERMISSIBLE CONCENTRATIONS[2149]

Radionuclide	Form	Concentration in Air (microcuries/milliliter)	Concentration in Water (microcuries/milliliter)
Iodine-129	Soluble	$2x10^{-11}$	$6x10^{-8}$
Iodine-129	Insoluble	$2x10^{-9}$	$2x10^{-4}$
Iodine-131	Soluble	$1x10^{-10}$	$3x10^{-7}$
Iodine-131	Insoluble	$1x10^{-8}$	$6x10^{-5}$
Krypton-85	Submersion*	$3x10^{-7}$	---
Xenon-133	Submersion*	$3x10^{-7}$	---

*Submersion means that the values given are for submersion in a semispherical infinite cloud of airborne material.

Option No.2-Activated Charcoal. Charcoal has been extensively used for the removal of elemental iodine from off-gas streams. Elemental iodine is readily trapped from air streams by activated charcoal, even at relative humidities approaching saturation. Iodine absorption efficiencies of 99.99 percent have been obtained.[1395] However, organic iodides, such as methyl iodide are not effectively absorbed. To remove organic iodides the activated-charcoal beds should be preceded by a catalytic oxidation step to completely oxidize all organic vapors in the gas stream. The addition of the oxidation step prior to the charcoal bed is effective for removing organic iodides, but the bed life may be limited to a few months or a residence time of a few seconds may be required which is not practical in a large gas flow system. The charcoal beds are quite effective for the removal of trace quantities of elemental iodine and their use should probably be limited to the final iodine removal step.

Option No.3-Impregnated Charcoal. Special impregnated charcoals have been developed for removing methyl iodide. The impregnated charcoals presently under consideration are: (1) iodized with one or more iodine-containing substances, to provide I-131, I-127 exchange capability; or (2) triethylinediamine - impregnated to effect removal by the utilization of the alkyl halide-organic amine reaction. Methyl iodine retention efficiencies greater than 99 percent have been obtained with both these impregnated charcoals.[0669] The removal efficiencies for the iodine-impregnated charcoal refer only to I-131. The performance of these impregnated charcoal beds is drastically reduced by poisoning due to impurities in the air stream.[1386] Iodine-impregnated charcoal and triethylinediamine-impregnated charcoal are satisfactory absorbents for organic iodides, such as methyl iodide, but an upstream filter system may be required to remove impurities in the air.

Option No.4-Silver Zeolite. Inorganic and organic forms of iodine are both effectively removed from the off-gas stream by silver zeolite. The silver zeolite has ideal properties of nonflammability, capability of iodine absorption up to 500 C, retention of absorbed iodine up to 1,000 C, and rapid reaction times. The zeolite is a synthetic zeolite absorber (Linde Molecular Sieve 13X) in which the normal sodium zeolite matrix has been converted to silver zeolite. The silver zeolite retention efficiency of elemental iodine and methyl iodide is greater than 99.8 percent at temperatures varying from room temperature to 500 C.[2291] The efficiency of the silver zeolite is adversely effected by long-term exposure to organic contaminants in the air stream. The zeolite life expectancy can be extended by preceding the zeolite with an oxidation step. This is economically desirable since the basic cost of silver zeolite is 3 to 10 times the cost of charcoal. Silver zeolite is the most efficient absorber of all airborne iodine species and is particularly applicable to the large gas flows encountered in nuclear fuel reprocessing plants.

Krypton and Xenon Recovery

Option No.1-Activated Charcoal. Krypton and xenon can be retained by adsorption beds of activated charcoal. In this process the krypton and xenon are cooled by means of cold trays and are then passed through a constant temperature (25 C) charcoal column. These absorption beds are used to provide holdup time which is long compared to the half-life of the isotopes. This process is effective for the 5.3 day xenon-133 but not for 10.8 year krypton-85. For xenon-133 the gas could be held up in a charcoal bed for a period of 18 days to reduce its activity 10 times or a period of 54 days for a reduction of a 1,000 times.

Option No.2-Cryogenic Distillation. A cryogenic distillation process for the recovery of krypton and xenon from off-gas streams generated during the processing of spent fuels has been applied at the Idaho Chemical Processing Plant.[2292] In this process the off-gas stream is first treated prior to entering the cryogenic system by passing the gas through a catalytic conversion system which removes nitrous oxide and hydrogen. In an off-gas stream with a high hydrogen content a special catalytic unit and condensor would also be required to reduce the hydrogen concentration in the off-gas to levels below the explosive limit. The off-gas is then fed to the cryogenic system which is cooled by liquid nitrogen. This system consists primarily of two regenerators, a distillation unit, and a batch still. The gas stream is cooled to -260 F in the two regenerators prior to entering the distillation column. In the distillation column, krypton and xenon are condensed and absorbed by liquid nitrogen which flows through the plates in the column. The krypton and xenon are dissolved in the liquid nitrogen and are transferred to a batch still where they are separated and purified in a fractionation step. This process has been applied in actual plant operation at the Idaho Chemical Processing Plant and is capable of processing up to 20 scfm of off-gas. The process has the potential for recovering 99 percent of the gases with 4 to 10 percent of nitrogen and oxygen impurities in the final product. Actual overall krypton-xenon recovery efficiencies have varied from 30 to 60 percent. The low recovery efficiencies were due to difficulties in operation of the gas cleanup step and to leaks within the product bottling system.

Option No.3-Fluorocarbon Solvents. Krypton and xenon can be effectively removed from contaminated air streams by selective absorption in fluorocarbon solvents, such as refrigerant-12 (dichlorodifluoromethane). This process has been tested on a pilot plant scale at processing rates up to 20 scfm of gas by Union Carbide at their Oak Ridge, Tennessee, facility.[1397] In this process, the removal of krypton and xenon is accomplished by contacting the gas stream with a stream of fluorocarbon solvent (refrigerant-11 or -12) in a packed absorber column at low temperatures (-177 to -21 F) and high pressure (169 to 437 psia). Besides krypton and xenon substantial amounts of oxygen, nitrogen, and argon are dissolved in the solvent. Since krypton and xenon are substantially more soluble in the solvent than these other gases, the absorber column is followed by a fractionator system where most of the dissolved nitrogen, argon, and oxygen is driven off. The solvent is then driven to a stripper system where the product gas concentrated in krypton and xenon is evolved and the solvent is recycled back to the absorber. Based on the pilot plant test data, a plant operating with an absorber at -30 F and 425 psia should give a krypton and xenon removal efficiency of over 99.7 percent with refrigerant-12 as the solvent.[1397] For application to fuel reprocessing plants with feed rates in excess of 100 scfm some scale-up of the pilot plant facility is required. This method offers the best potential for the effective removal of krypton and xenon from off-gas streams. It has been successfully operated on a pilot-plant scale but has not yet been applied in actual plant operations.

Storage/Disposal

Option No.1-Land Burial. Land burial of iodine, krypton, and xenon wastes, in small concentrations, at approved sites that are acceptable from a geologic and hydrologic standpoint, is an acceptable means of disposal. Their concentrations should not be in excess of 10^4 times their maximum permissible concentrations for the general population in 10CFR20.[2149] The burial trenches should be designed not to intercept the ground water table and constructed with a bottom drain and sump for water monitoring. The trenches should be covered with either asphalt or vegetation to limit infiltration of water. The burial site design, geology, and hydrology should be in conformance with the criteria used in selecting and licensing the present commercial burial sites.[1423] Since land burial has successfully been practiced on a commercial scale, it should be considered as the most satisfactory method of disposing of low concentrations of these wastes.

Option No.2-Near-Surface Storage. The storage of high concentrations of these wastes in engineered storage facilities offers the best immediate method for storage of these wastes. The technology for these facilities has been developed. The wastes will be under surveillance and control and can be retrieved, should this be required. The wastes will be stored in stainless-steel lined concrete vaults which will be either air or water cooled.

Option No.3-Salt Deposits. This method offers the best potential for the disposal of this radionuclide. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November of 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive wastes is satisfactory. 0733 Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as an ultimate method of disposal. The wastes are buried in rooms carved in the salt deposits approximately 1,000 ft below the ground. The salt is a good heat transmitter, provides about the same radioactive shielding as concrete and can heal its own fractures by plastic flow. The critical problem is the selection of a site that meets the necessary design and geological criteria.

To summarize, krypton, xenon, and iodine are released in the off-gas streams during processing of the spent fuels. The iodine can be removed from the off-gas stream by silver zeolite. Krypton and xenon can be effectively removed by cryogenic distillation or by selective absorption in fluorcarbon solvents. Following recovery the wastes can be held to allow the short-lived isotopes such as iodine-131 and xenon-133 to decay. For iodine-131 and xenon-133 their activity is reduced by a factor of 10,000 in 106 days and 71 days, respectively. The long-lived isotopes krypton-85 (10.8 years) and iodine-129 (17 million years) should be encapsulated and disposed of in salt deposits following storage in near-surface engineered facilities. The wastes to be disposed of can be encapsulated in high-pressure cylinders or solidified. A method for solidifying the gases xenon and krypton includes dispersion in glasses or resins and entrapment in molecular sieves or small pressurized steel bulbs which are in turn encased in epoxy resin.

6. APPLICABILITY TO NATIONAL DISPOSAL SITES

Iodine-129 and krypton-85 are candidates for a National Disposal Site due to their long half-lives and projected growth with that of the civilian nuclear power program. Iodine-131 and xenon-133 are not candidates for a

National Disposal Site because of their short half-lives.

The recommended treatment for the recovery of iodine-129 and iodine-131 from reprocessing plant off-gas streams is by the use of silver zeolite. Krypton-85 and zenon-133 can be removed by cryogenic distillation or absorption in fluorocarbon solvents. Iodine-131 and xenon-133 can be stored in engineered storage facilities to allow their radioactive decay and then disposed of in low concentrations in land burial facilities. Iodine-129 and xenon-133 should be stored in engineered storage facilities followed by disposal in salt beds.

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HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Iodine-129

Half-life $1.7\text{x}10^7$ years

Type of Decay Negative Beta

Structural Formula

$_{53}I^{129}$

Molecular Wt. 129 Melting Pt. 114 C Boiling Pt. 184 C

Density Gas: 11.27 grams/liter
Solid: 4.93 gm/cc at 20 C

Specific Power $7.08\text{x}10^{-8}$ watts/gm

Specific Activity $1.62\text{x}10^{-4}$ curies/gm

Solubility

Cold Water Slightly soluble Hot Water Slightly soluble

Others: Dissolves readily in chloroform, carbon tetrachloride, & carbon disulfide

Decay Chain

$I^{129} \longrightarrow Xe^{129}$
($1.7\text{x}10^7$y) (Stable)

Radiation Energy Level & Intensities

Beta: 0.150 mev (100%)
Gamma: 0.038 mev (100%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Iodine-131

Structural Formula $_{53}I^{131}$

Half-life 8 days

Type of Decay Negative Beta

Molecular Wt. 131 Melting Pt. 114 C Boiling Pt. 1840 C

Density Gas: 11.27 gm/liter
Solid: 4.93 gm/cc at 20 C

Specific Power -

Specific Activity 1,230 curies/gm

Solubility

Cold Water Slightly soluble Hot Water Slightly soluble

Others: Dissolves readily in chloroform, carbon tetrachloride,& carbon disulfide

Decay Chain

$$\underset{(8d)}{I^{131}} \longrightarrow \underset{(stable)}{Xe^{131}}$$

Radiation Energy Level & Intensities

Beta:	0.810 Mev	(0.7%)
	0.608 Mev	(82.7%)
	0.330 Mev	(9.3%)
	0.250 Mev	(2.8%)
Gamma:	0.723 Mev	(1.6%)
	0.637 Mev	(6.8%)
	0.364 Mev	(82%)
	0.284 Mev	(5.4%)
	0.080 Mev	(2.6%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Xenon-133

Half-life 5.3 days

Type of Decay Negative Beta

Structural Formula

$\boxed{_{54}Xe^{133}}$

Molecular Wt. 133 Melting Pt. -112 C Boiling Pt. -107 C

Specific Power ________

Specific Activity 1.86×10^{5} curies/gm

Solubility

Cold Water ________ Hot Water ________

Others: Chemically inert

Density

Gas:	5.495 gm/liter at 20 C
Liquid:	3.52 gm/cc at -107C
Solid:	2.70 gm/cc at

Decay Chain

$$\underset{(5.3\ \text{days})}{Xe^{133}} \xrightarrow{B^{-1}} \underset{(\text{stable})}{Cs^{133}}$$

Radiation Energy Level & Intensities

Beta: 0.346 Mev (100%)

Gamma: 0.809 Mev (100%)

Shipping Regulations Classified as a transport group VI radionuclide at pressures less than 14.7 psia and as a group III radionuclide at pressures greater than 14.7 psia by the Department of Transportation

Comments Xenon is a member of the so-called rare or inert gases

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Krypton-85

Half-life 10.8 years

Tyne of Decay Negative Beta

Molecular Wt. 85 Melting Pt. -157 C Boiling Pt. -152 C

Density Gas: 3.733 gm/liter @ 0 C
Liquid: 2.16 gm/cc @ -153 C

Specific Power 0.53 watts/gm

Specific Activity 397 curies/gm

Structural Formula

$_{36}Kr^{85}$

Solubility

Cold Water ______ Hot Water ______

Others: Chemically inert

Decay Chain

$$\underset{(10.8y)}{Kr^{85}} \xrightarrow{B^{-1}} \underset{(stable)}{Rb^{85}}$$

Radiation Energy Level & Intensities

Beta: 0.690 Mev (99.4%)
0.150 Mev (0.6%)

Gamma: 0.514 Mev (0.4%)

Shipping Regulations Classified as a transport group VI radionuclide at pressures less than 14.7 psia and as a group III radionuclide at pressures greater than 14.7 psia by the Department of Transportation

Comments Krypton is a member of the so-called rare or inert gases.

References: (1) 0766
(2) 2150

PROFILE REPORTS

Plutonium-238, Plutonium-239, Plutonium-240, Plutonium-241 Americium-241, Americium-243, Curium-242, Curium-244

1. GENERAL

Introduction

Plutonium, americium, and curium are artificially produced radionuclides that do not exist in nature except for very small amounts of plutonium. These elements are characterized by their high radiotoxicity, long half-life, and ability to fission. Plutonium-239 is the most important among these elements because of its use in nuclear weapons and the place it holds as the key material in the development of fast-breeder reactors for the civilian nuclear power program. Plutonium-239 is readily fissionable with neutrons and one lb of this material is equivalent to about ten billion watt-hours of heat energy.

Americium, curium, and plutonium (except for Pu-241) decay by the emission of high energy, 5 to 6 Mev, alpha particles. Plutonium-241 decays by beta emission to form americium-241. All these elements eventually decay to lead, and the time required is on the order of millions of years. The americium-243 decay chain is attached (Table 1) and the three long-lived isotopes in this chain are: Pu-239, half-life 2.4×10^4 years; U-235, half-life 7.1×10^8 years; and Pu-231, half-life 3.3×10^4 years.

The projected growth in production of these isotopes will parallel that of the civilian nuclear power program. The production of plutonium-239 will increase with the planned introduction of the fast-breeder reactors in the 1980's. Present projections indicate that nuclear power will account for 30 percent of the total power production by the year 1980.[2148] The expected total

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TABLE 1

AMERICIUM-243 DECAY CHAIN

Nuclide	Name	Half-Life	Major Radiation
${}_{95}\mathrm{Am}^{243}$	Americium-243	7,950 years	Alpha and gamma
${}_{93}\mathrm{Np}^{239}$	Neptunium-239	2.4 days	Beta and gamma
${}_{94}\mathrm{Pu}^{239}$	Plutonium-239	24,400 years	Alpha and gamma
${}_{92}\mathrm{U}^{235}$	Uranium-235	$7.1\mathrm{x}10^{8}$years	Alpha and gamma
${}_{90}\mathrm{Th}^{231}$	Thorium-231	25 hours	Beta and gamma
${}_{91}\mathrm{Pa}^{231}$	Protactinium-231	$3.3\mathrm{x}10^{4}$ years	Alpha and gamma
${}_{89}\mathrm{Ac}^{227}$	Actinium-227	21.6 years	Beta and gamma
${}_{90}\mathrm{Th}^{227}$	Thorium-227	18.2 days	Alpha and gamma
${}_{88}\mathrm{Ra}^{223}$	Radium-223	11.4 days	Alpha and gamma
${}_{86}\mathrm{Rn}^{219}$	Radon-219	4 seconds	Alpha and gamma
${}_{84}\mathrm{Po}^{215}$	Polonium-215	1.8 minutes	Alpha
${}_{82}\mathrm{Pb}^{211}$	Lead-211	36 minutes	Beta and gamma
${}_{83}\mathrm{Bi}^{211}$	Bismuth-211	2 minutes	Alpha,beta,and gamma
${}_{81}\mathrm{Tl}^{207}$	Thallium-207	4.8 minutes	Beta and gamma
${}_{82}\mathrm{Pb}^{207}$	Lead-207	Stable	

accumulated wastes for these isotopes from the civilian nuclear power program to the year 2020 is attached (Table 2). These data assume that 99.5 percent of plutonium is recovered from the spent fuel elements. Annual production figures or accumulated totals for these materials produced as part of the nuclear weapons program were not available.

Manufacture

The transuranium elements are produced in cyclotrons and in thermal power reactors. The majority of the isotopes are produced in civilian nuclear power plants. At the present time there are 22 nuclear power plants in operation with an additional 104 being built or planned.[2151] The present reactor fuel is uranium dioxide. In the future, plutonium dioxide will be used as the fuel in the fast-breeder reactors.

Plutonium. Plutonium-238 is produced in cyclotrons by deuteron bombardment of uranium or in nuclear reactors from neutron bombardment by the reaction:

$$U^{238}(n,2n)U^{237} \xrightarrow{B^-} Np^{238} \xrightarrow{B^-} Pu^{238}$$

Plutonium-238 is also a daughter of Cm-242. Plutonium-239 is produced in extensive quantities in nuclear reactors from natural uranium by the reaction:

$$U^{238}(n, \gamma)U^{239} \xrightarrow{B^-} Np^{239} \xrightarrow{B^-} Pu^{239}$$

Plutonium-240 and plutonium-241 are produced by multiple n-capture from U-238 and Pu-239.

Americium. Americium-241 is a daughter of plutonium-241 and is formed from plutonium-239 in a nuclear reactor by successive neutron capture. Americium-243 is produced by multiple n-capture from U-238 and Pu-239.

TABLE 2

ACCUMULATED ACTINIDES IN SPENT FUEL PROCESSING WASTES FROM THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Accumulated Radionuclides megacuries	Calendar Year 1970	1980	1990	2000	2020
Plutonium-238*	0.002	1.2	8.3	30.7	166
Plutonium-239*	0.00009	0.02	0.24	1.3	8.5
Plutonium-240*	0.0001	0.04	0.4	1.9	11.4
Plutonium-241*	0.03	6.6	47.2	191	909
Americium-241	0.009	2.3	22.7	121	763
Americium-243	0.0009	0.23	1.5	5.2	27.0
Curium-242	0.73	43.2	185.0	487.0	1,490
Curium-244	0.13	29.9	137.0	255.0	700

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*Assumes that 0.5 percent of the plutonium in the spent fuel is lost to waste.

Curium. Curium-242 is produced in cyclotrons by the helium-ion bombardment of Pu-239. It is also produced by multiple n-capture from U-238 and Pu-238 and by the reaction:

$$Am^{241}(n,\gamma)Am^{242} \xrightarrow{B^-} Cm^{242}$$

Cerium-244 is produced by multiple n-capture from U-238, Pu-239, and Am-243.

Uses

Plutonium-238 is used extensively for radioisotopic power devices for space electric power, for radioisotopic heaters, and as an energy source for cardiac pacemakers, heart pumps, and small undersea propulsion systems. Plutonium-239 is used as a fuel in fast-breeder reactors and in nuclear weapons. Presently, plutonium-239 is used in experimental reactors but with the introduction of the fast-breeder reactors in the civilian nuclear power program the requirements for its use will increase rapidly. Curium and americium are used in various national laboratories for research purposes and to produce heavier elements.

Sources and Types of Wastes

The majority of the transuranium elements are produced in nuclear reactors and are contained within the spent fuel elements. The spent fuel elements are processed for the recovery of the usable fission materials (uranium and plutonium) which are refabricated into new fuel elements. Solvent extraction using nitric acid is the means currently used for the first-stage removal of the fissionable materials. Approximately 99 percent of the plutonium is removed during the processing. The other radionuclides along with the unrecovered plutonium are contained in the aqueous effluent from the reprocessing step. The composition of fuel elements prior to processing have been tabulated,[0705] and the activity of plutonium, americium, and curium products present in the waste stream following the processing step are included (Table 3).

TABLE 3
ACTIVITY OF PLUTONIUM, AMERICIUM, AND CURIUM PRODUCTS PRESENT IN WASTES GENERATED BY THE PROCESSING OF SPENT REACTOR FUELS0705*

Light Water Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Actinide Products	After 1 year Grams/Tonne	After 1 year Watts/Tonne	After 1 year Curies/Tonne	After 100 years Grams/Tonne	After 100 years Watts/Tonne	After 100 years Curies/Tonne
Pu^{238}, Pu^{239}, Pu^{240}, Pu^{241}	48	2.7	641	71	2	64
Am^{241}, Am^{243}	144	6.2	189	140	6	179
Cm^{242}, Cm^{244}	32	306	8,430	1	2	57
Other Actinides	5,436	0.1	30	5,438	0.1	25

Fast-Breeder Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Actinide Products	After 1 year Grams/Tonne	After 1 year Watts/Tonne	After 1 year Curies/Tonne	After 100 years Grams/Tonne	After 100 years Watts/Tonne	After 100 years Curies/Tonne
Pu^{238}, Pu^{239}, Pu^{240}, Pu^{241}	428	11	3,203	412	8	264
Am^{241}, Am^{243}	742	54	1,620	794	48	1,469
Cm^{242}, Cm^{244}	20	625	17,000	1	3	72
Other Actinides	4,550	2	277	4,637	1	165

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*Assumes that 0.5 percent of the plutonium in the spent fuel is lost to waste.

Physical and Chemical Properties

The physical and chemical properties of plutonium, americium, and curium are included in the attached worksheets. Plutonium is highly reactive, and in moist air oxidation proceeds rapidly, especially at elevated temperatures. It dissolves in concentrated hydrochloric acid, hydroiodic acid, or perchloric acid with the formation of the Pu^{+3} ion. Plutonium exhibits six allotropic modifications having various crystalline structures whose densities vary from 15.92 to 19.84. Plutonium forms binary compounds with oxygen, carbon, nitrogen, silicon, and the halides.

Americium is a ductile, malleable metal which is precipitated by the fluorides, hydroxides, and oxalates. The element exists in three oxidation states in aqueous solution. The trivalent state is highly stable and difficult to oxidize. Curium is chemically similar to gadolenium and is carried on rare-earth precipitates. The only stable oxidation state that has been definitely identified in aqueous solutions is +3.

2. RADIATION HAZARD

Plutonium is one of the most dangerous poisons known. The permissible levels of concentration of plutonium in air and water are the lowest of any of the radioactive elements. This is a result of the concentration of plutonium directly in the blood-forming sections of the bone, rather than the more uniform bone distribution shown by other heavy elements. Americium and curium are also extremely hazardous radioactive materials.

The effects of their radiation exposure are primarily dependent on the amount of radiation and the portion of the body affected. The dose delivered to a particular body organ following the inhalation of 1 microcurie of each of these radionuclides is attached (Table 4). For plutonium-239 the dose delivered to the bone is 7,000 rem following the

TABLE 4

PLUTONIUM, AMERICIUM, AND CURIUM DOSE TO A PARTICULAR BODY ORGAN FOLLOWING INHALATION OF ONE MICROCURIE OF THE NUCLIDE

Isotope	Form	Organ	Dose rem
Plutonium-238	Insoluble	Lung	64
Plutonium-238	Soluble	Bone	700
Plutonium-239	Insoluble	Lung	60
Plutonium-239	Soluble	Bone	7,000
Plutonium-240	Insoluble	Lung	60
Plutonium-240	Soluble	Bone	7,000
Plutonium-241	Insoluble	Lung	0.06
Plutonium-241	Soluble	Bone	130
Americium-241	Insoluble	Lung	65
Americium-241	Soluble	Bone	2,200
Americium-243	Insoluble	Lung	60
Americium-243	Soluble	Bone	2,000
Curium-242	Insoluble	Lung	43
Curium-242	Soluble	Liver	50
Curium-244	Insoluble	Lung	67
Curium-244	Soluble	Bone	1,300

inhalation of 1 microcurie (16.4 micrograms). The dose delivered to the bone following the injection of 1 microcurie into the body via a wound is 30,000 rem.

Standards for prolonged exposure over a fifty-year period have defined the single dose limit in terms of the maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter and the accumulated dose limit is 5(N - 18), where N is the individual's age in years. Limits for the thyroid, bone, and other organs have also been defined. Values of the total body burden for each radionuclide required to produce the maximum permissible dose rates defined above have been compiled.[0563] For plutonium the critical organ is the bone and the maximum permissible body burden is 0.04 microcuries for Pu-238, Pu-239, and Pu-240 and 0.9 microcuries for Pu-241. For americium-241 and americium-243 the critical organ is the bone and the maximum permissible body burden is 0.05 microcuries. For curium-242 the critical organ is the liver and the maximum body burden is 0.05 microcuries. For curium-244 the critical organ is the bone and the maximum body burden is 0.1 microcuries.

3. OTHER HAZARDS

Besides its radiation hazard, plutonium is highly toxic. It is also highly reactive in moist air. The toxicity of curium and americium are unknown.[0766] Plutonium, americium, and curium also present a potential hazard due to nuclear fission. Plutonium-239 represents the greatest hazard since it readily undergoes fission with thermal neutrons, as well as with those of higher energy and is capable of maintaining a self-sustaining reaction.

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

These radionuclides are all alpha emitters except for plutonium-241. The alpha particles have little penetrating power and can be stopped by a sheet of writing paper. They are extremely hazardous if inhaled. Semi-remote handling techniques are required to protect personnel from radiation injury and adequate radiation shielding is required to avoid excessive radiation exposure. These materials are safely handled in glove boxes. The beta particles emitted by plutonium-241 are of low energy (0.021 Mev) and cannot penetrate more than 0.002 in. of water.

These radionuclides are stored in controlled areas in specially constructed containers. Extreme precautions are taken to avoid the release of any material. Plutonium is stored at low temperatures and in dry air to avoid corrosion. The storage container is often protected by both a primary and a secondary containment barrier. Precautions are also taken to prevent the formation of a critical mass. Plutonium in a liquid solution is more likely to become critical than solid plutonium. The container design and arrangement must preclude any possibility of obtaining nuclear criticality. Special monitoring systems and proper warning signs are located in the general area of the storage facility.

Plutonium, americium, and curium are classified as a transport group I radionuclide by the Department of Transportation, and the rules and regulations governing their transportation are given in the Code of Federal Regulations (CFR) Title 49--Transportation, Parts 170 to 190.[2150] Their content is limited to 0.001 curies for a Type A package and 20 curies for a Type B package defined in 49CFR173. The limits are increased to 20 and 500 curies if their physical form meets the requirements of a special form material. Their release rate is limited to zero under the specified accident conditions for Type A and B quantities.[2150] Plutonium-239 is designated as a fissile material by the Department of Transportation and must meet the controls required to provide nuclear criticality safety.

Disposal/Reuse

The disposal of these materials is governed by the AEC Manual Chapter 0524[0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The standards for the safe release of these materials to the environment in an unrestricted area are contained in 10CFR20[2149] and their concentrations in air and water should not exceed the values listed in this report (Table 5). These concentrations apply to an individual and their release may be limited if a suitable sample of the population is exposed to one-third the concentrations in air or water specified in this report (Table 5).

Although not practiced, the disposal by release in a sanitary sewage system is limited to 0.1 microcurie.[2149] The disposal by burial at any one location and time is limited to 10 microcuries.[2149]

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Recovery

Option No. 1 - Solvent Extraction. Solvent extraction processes have successfully been used for well over a decade as the standard method for the recovery of up to 99.8 percent of the plutonium and uranium in spent reactor fuel elements. The basis for separation in this process is the differences in solubility of these materials in the organic and aqueous phase. Nearly all major fuel reprocessing facilities use the Purex solvent extraction process. These facilities include those operated under contract to the Atomic Energy Commission (AEC) and Nuclear Fuel Services facility at West Valley, New York, which is privately owned and operated. In the Purex process, the spent fuel elements are dissolved in nitric acid and an organic compound, tributyl phosphate (TBP) in an inert hydrocarbon, is added. When this organic mixture is added, the TBP extracts both the uranium and the plutonium

TABLE 5

PLUTONIUM, AMERICIUM, AND CURIUM MAXIMUM PERMISSIBLE CONCENTRATIONS[2149]*

Radionuclide	Form	Concentration in Air (microcuries/milliliter)	Concentration in Water (microcuries/milliliter)
Plutonium-238	Soluble	7×10^{-14}	5×10^{-6}
Plutonium-238	Insoluble	1×10^{-12}	3×10^{-5}
Plutonium-239	Soluble	6×10^{-14}	5×10^{-6}
Plutonium-239	Insoluble	1×10^{-12}	3×10^{-5}
Plutonium-240	Soluble	6×10^{-14}	5×10^{-6}
Plutonium-240	Insoluble	1×10^{-12}	3×10^{-5}
Plutonium-241	Soluble	3×10^{-12}	2×10^{-4}
Plutonium-241	Insoluble	1×10^{-9}	1×10^{-3}
Americium-241	Soluble	2×10^{-13}	4×10^{-6}
Americium-241	Insoluble	4×10^{-12}	2×10^{-5}
Americium-243	Soluble	2×10^{-13}	4×10^{-6}
Americium-243	Insoluble	4×10^{-12}	3×10^{-5}
Curium-242	Soluble	4×10^{-12}	2×10^{-5}
Curium-242	Insoluble	6×10^{-12}	3×10^{-5}
Curium-244	Soluble	3×10^{-13}	7×10^{-6}
Curium-244	Insoluble	3×10^{-12}	3×10^{-5}

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*Maximum permissible concentrations apply to an individual in an unrestricted area.

into the organic phase, while the fission products and other wastes remain in the aqueous phase. Plutonium and uranium are separated by reducing the plutonium to the trivalent form, usually with ferrous sulfamate, and contacting with nitric acid. In the aquafluor process, to be used at General Electric's Morris, Illinois plant, the uranium and plutonium separation will be accomplished by ion exchange. The ion exchange separation step will limit the plant's capacity since they must be designed to prevent the accumulation of a critical mass.

Storage/Disposal

Option No. 1 - Land Burial. Disposal of these high-level radionuclides, in small concentrations, by direct burial in unlined trenches is not considered a satisfactory means of disposal. These types of wastes are presently disposed of in this manner at approved sites. This method of disposal does not provide for any type of secondary containment barrier to prevent their release by leaching with the local ground water. Since these materials have an extremely long half-life and a high radiotoxicity, this method of disposal is not satisfactory.

Option No. 2 - Near-Surface Liquid Storage. Near-surface storage of these radionuclides in stainless steel tanks encased in concrete and buried underground is not considered as a satisfactory means of disposal. Aqueous solutions of spent fuel reprocessing wastes have been stored in this manner over the past 25 years. These tanks are considered as an interim storage technique due to a general lack of confidence in their long-term integrity.[0705] Therefore, the near-surface storage of these wastes in steel tanks should only by considered as an interim storage technique and not as a permanent storage or disposal technique.

Option No. 3 - Near-Surface Solid Storage. The storage of solidified wastes in engineered surface facilities offers the best immediate method for storage of these wastes. The necessary technology for these facilities has been developed. The wastes will be under constant surveillance and control and can be retrieved, should this be required. The wastes should be solidified and encapsulated in a suitable container (steel). Of the four high-level solidification processes developed for reactor-produced wastes, spray or phosphate glass solidification offer the better solidified waste characteristics than the pot calcination or fluidized bed calcination processes (see Radioactive Waste Solidification report). The wastes will be stored in stainless steel-lined concrete vaults which will be either air or water cooled.

Option No. 4 - Salt Deposits. This method offers the best potential for the disposal of solidified, high-level wastes since bedded salt deposits are completely free of circulating ground waters. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November of 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive waste is satisfactory.[0733] Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as an ultimate method of disposal. The wastes must be solidified and disposed of in the solid form encapsulated in a suitable container. The solidified wastes are buried in rooms carved in the salt deposits approximately 1,000 ft below the ground. The salt is a good heat transmitter, provides about the same radioactive shielding as concrete, and can heal its own fractures by plastic flow. The critical problem is the selection of a site that meets the necessary design and geological criteria for the mixture of fission products and actinide wastes.

Option No. 5 - Bedrock Disposal. The disposal of high-level liquid wastes along with other spent fuel processing wastes in vaults excavated in crystalline rock over 1,500 ft beneath the ground is currently being evaluated by E. I. Du Pont de Nemours, at their Savannah River Plant near Aiken, South Carolina.[0894,1396] An advisory committee appointed by the National Academy of Sciences recommended abandonment of the project. In May 1972, another National Academy of Sciences panel concluded that bedrock storage provides a reasonable prospect for long-term safe storage, but precise information is needed to decide if and where underground storage vaults should be built. Another method has also been proposed for disposing of liquid wastes by in situ incorporation in molten silicate rock.[2146] At the present time both of these methods are unproven since sufficient engineering data or exploration has not been completed to verify their suitability. Due to the long half-lives and high biological hazard of the actinides, it is doubtful that data could ever be accumulated to prove that the geological characteristics of the site over the next few thousand years are acceptable to ensure the absolute safety of such a disposal method.

To summarize, plutonium, americium, and curium are hazardous, long-lived isotopes for which extreme precautions are required in their disposal. Plutonium is separated from the spent fuel processing wastes by solvent extraction for reuse. Recovery factors as high as 99.8 percent can be obtained. The americium, curium, and remaining plutonium are contained with the final reprocessing wastes. The acceptable methods of treatment are spray or phosphate glass solidification and disposal in either salt deposits or near-surface engineered facilities. Plutonium, americium, and curium wastes from research laboratories and other sources should also be solidified, encapsulated, and disposed of in the same manner.

6. APPLICABILITY TO NATIONAL DISPOSAL SITE

Plutonium, americium, and curium are candidates for a National Disposal Site due to their long half-lives, health hazard, and projected growth with that of the civilian nuclear power program. From a cost and safety viewpoint, it would be desirable to combine the reprocessing plant and the National Disposal Site. Interim storage in near-surface, engineered storage facilities offers considerable latitude in the site selection. The site selection for ultimate disposal of these wastes is limited to particular geological areas in which salt deposits are present.

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HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Plutonium-238

Structural Formula

$_{94}Pu^{238}$

Half-life 86.4 years

Type of Decay Alpha and Spontaneous Fission

Molecular Wt. 238 Melting Pt. 639 C Boiling Pt. 3235 C

Density 19.84 gm/cc Specific Power 0.56 watts/gm

Specific Activity 17.4 curries/gm

Solubility

Cold Water ______ Hot Water ______

Others: Soluble HCl; insoluble in HNO_3 and concentrated H_2SO_4

Decay Chain

$$\underset{(86.4y)}{_{94}Pu^{238}} \xrightarrow{\alpha} \underset{(2.5X10^5y)}{_{92}U^{234}}$$

Radiation Energy Level & Intensities

Alpha: 5.499 Mev (72%)
5.456 Mev (28%)

Gamma: 0.0435 Mev (0.04%)

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation

Comments ______

References:
(1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Plutonium-239

Half-life 24,400 years

Type of Decay Alpha and Spontaneous Fission

Structural Formula

$_{94}Pu^{239}$

Molecular Wt. 239 Melting Pt. 639 C Boiling Pt. 3235 C

Density 19.84 gm/cc Specific Power 0.0019 watts/gm

Specific Activity 0.061 curries/gm

Solubility

Cold Water ____ Hot Water ____

Others: Soluble HCl, insoluble in HNO_3 and concentrated H_2SO_4

Decay Chain

$$\underset{(2.4\times10^4 y)}{_{94}Pu^{239}} \xrightarrow{\alpha} \underset{(7.1\times10^8 y)}{_{92}U^{235}}$$

Radiation Energy Level & Intensities

Alpha:	5.157 Mev	(74%)
	5.145 Mev	(15%)
	5.107 Mev	(11%)
Gamma:	0.414 Mev	(0.0001%)
	0.375 Mev	(0.0001%)
	0.129 Mev	(0.005%)
	0.052 Mev	(0.020%)
	0.039 Mev	(0.007%)

Shipping Regulations ____

Comments Classified as a transport group I radionuclide by the Department of Transportation

References: (1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Plutonium-240

Structural Formula

$_{94}Pu^{240}$

Half-life 6,580 years

Type of Decay Alpha and Spontaneous Fission

Molecular Wt. 240 Melting Pt. 639 C Boiling Pt. 3235 C

Density 19.84 gm/cc Specific Power 0.0069 watts/gm

Solubility Specific Activity 0.227 curries/gm

Cold Water Hot Water

Others: soluble HCl; insoluble in HNO_3 and concentrated H_2SO_4

Decay Chain

$$\underset{(6580y)}{_{94}Pu^{240}} \xrightarrow{\alpha} \underset{(2.4X10^7y)}{_{92}U^{236}}$$

Radiation Energy Level & Intensities

Alpha: 5.168 Mev (76%)
5.123 Mev (29%)

Gamma: 0.045 Mev (24%)

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Plutonium-241

Half-life 13.2 years

Type of Decay Negative Beta

Structural Formula

${}_{94}Pu^{241}$

Molecular Wt. 241

Melting Pt. 639 C

Boiling Pt. 3235 C

Density 19.84 gm/cc

Specific Power 0.0048 watts/gm

Specific Activity 112 curries/gm

Solubility

Cold Water

Hot Water

Others: Soluble HCl; insoluble in HNO_3 and concentrated H_2SO_4

Decay Chain

$${}_{94}Pu^{241} \xrightarrow{B^-} {}_{95}Am^{241}$$

(13.2y) (458y)

Radiation Energy Level & Intensities

Beta: 0.021 Mev (100%)

Also emits alpha particle 0.0023% of the time.

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation.

Comments

References:
(1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Americium-241

Half-life 458 years

Type of Decay Alpha and Spontaneous Fission

Structural Formula $_{95}Am^{241}$

Molecular Wt. 241 Melting Pt. 994 C Boiling Pt. 2607 C

Density 11.7 gm/cc Specific Power 0.11 watts/gm

Solubility Specific Activity 3.24 curries/gm

Cold Water Hot Water

Others: Soluble in dilute acid

Decay Chain

$$_{95}Am^{241} \xrightarrow{\alpha} {}_{93}Np^{237}$$

(458y) (2X10^{6}y)

Radiation Energy Level & Intensities

Alpha: 5.486 Mev (86%)
5.443 Mev (12.7%)
5.389 Mev (1.3%)

Gamma: 0.060 Mev (36%)
0.026 Mev (3%)

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation.

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Americium-243

Half-life 7,950 years

Type of Decay Alpha

Molecular Wt. 243

Melting Pt. 994 C

Boiling Pt. 2607 C

Density 11.7 gm/cc

Specific Power 0.0062 watts/gm

Specific Activity 0.185 curries/gm

Structural Formula

$_{95}Am^{243}$

Solubility

Cold Water

Hot Water

Others: Soluble in dilute acid

Decay Chain

$_{95}Am^{243} \xrightarrow{\alpha} {}_{93}Np^{239} \xrightarrow{B^-} {}_{94}Pu^{239}$

(7,950y) (2.4d) ($2.4X10^4$y)

Radiation Energy Level & Intensities

Americium-243

Alpha: 5.276 Mev (88%)
5.234 Mev (11%)
5.181 Mev (1.1%)

Gamma: 0.075 Mev (50%)
0.044 Mev (4%)

Neptunium-239

Beta: 0.437 Mev (Max)

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation.

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Curium-242

Half-life 163 days

Type of Decay Alpha and Spontaneous Fission

Molecular Wt. 242   Melting Pt. 1340 C   Boiling Pt. ____

Density 7 gm/cc   Specific Power 122 watts/gm

Structural Formula

$_{96}\mathrm{Cm}^{242}$

Solubility   Specific Activity 3,320 curries/gm

Cold Water ____   Hot Water ____

Others: ____

Decay Chain

$$_{96}\mathrm{Cm}^{242} \xrightarrow{\alpha} {}_{94}\mathrm{Pu}^{238}$$

(163d)   (86 years)

Radiation Energy Level & Intensities

Alpha: 6.115 Mev (74%)
6.071 Mev (26%)

Gamma: 0.158 Mev (.003%)
0.102 Mev (.004%)
0.044 Mev (.041%)

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation.

Comments ____

References: (1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Curium-244

Half-life 17.6 years

Type of Decay Alpha and Spontaneous Fission

Molecular Wt. 244

Melting Pt. 1340 C

Boiling Pt.

Density 7 gm/cc

Structural Formula

$_{96}Cm^{244}$

Specific Power 2.7 watts/gm

Specific Activity 83 curries/gm

Solubility

Cold Water

Hot Water

Others:

Decay Chain

$$_{96}Cm^{244} \xrightarrow{\alpha} {}_{92}Pu^{240}$$

(17.6y) (6580y)

Radiation Energy Level & Intensities

Alpha: 5.806 Mev (77%)
5.766 Mev (23%)

Gamma: 0.150 Mev (.0013%)
0.100 Mev (.0015%)
0.043 Mev (.0210%)

Shipping Regulations Classified as a transport group I radionuclide by the Department of Transportation.

Comments

References: (1) 0766
(2) 2147
(3) 2156

PROFILE REPORT

Ruthenium-106 (Rhodium-106), Cerium-144 (Praseodymium-144), Promethium-147

1. GENERAL

Introduction

Ruthenium-106, cerium-144, and promethium-147 are radioactive isotopes that are produced in nuclear reactors by the fission of uranium. Ruthenium-106 has a half-life of 368 days and emits only beta particles. It exists in radiation equilibrium with its short half-lived daughter, rhodium-106. Rhodium-106 has a half-life of 30 seconds and emits both gamma rays and high energy (3.55 Mev) beta particles. Cerium-144 is both a beta and gamma emitter and has a half-life of 284 days. It decays by beta emission to form praseodymium-144 which has a half-life of 17.3 minutes. Promethium-147 has a half-life of 2.62 years and decays by the emission of a 0.224 beta particle to form stable samarium-147.

These three isotopes, Ru-106, Ce-144, and Pm-147 have moderately long half-lives (284 days to 2.6 years) and account for a majority of the total fission product activity and heat content in the spent fuel processing. After one year of radioactive decay, they are responsible for 65 percent of the total heat content and for 70 percent of the total activity in the high-level waste.

Since these isotopes are principally produced in nuclear reactors, their projected growth will parallel that of the civilian nuclear power program. Present projections indicate that nuclear power will account for 30 percent of the total power production by the year 1980.[2148] Annual production figures from the civilian and nuclear power program for these three radionuclides to the year 2020 for light-water and fast-breeder reactor fuels are attached (Tables 1 to 3). These figures illustrate the tremendous growth anticipated in their production.

Manufacture

Presently, these three radionuclides are produced in thermal reactors by the fission of uranium. The fission yield for these three radionuclides is: cerium-144, 5.6 percent; promethium-147, 2.6 percent; and ruthenium-106, 0.38 percent. Civilian nuclear power plants are the major producers and a smaller amount is produced at AEC facilities. At the present time there are 22 nuclear power plants in operation with an additional 104 being built or planned.[215] They will also be produced in fast-breeder reactors by the fission of plutonium.

Uses

Promethium-147 is used in radioisotope power generators and as a beta source in the radioactive gauge field. It is also used in the preparation of self-illuminating materials and devices for signs and signals. At the present time promethium-147 is separated from the reactor-produced waste streams and distributed by the AEC through the Oak Ridge National Laboratory, Isotope Sales Department. The AEC revenue from its distribution for the years 1968 to 1971 is[215]:

Year	Revenue (Dollars)
1968	56,000
1969	45,000
1970	16,000
1971	30,000

In the future when its production is warranted by market demands, it would be available in large quantities from commercial firms operating chemical reprocessing plants. The commercial use for ruthenium-106 and cerium-144 is very limited.

TABLE 1

RUTHEMIUM-106 CONTENT IN HIGH-LEVEL WASTE PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year	$.071x10^{5}$	$4.06x10^{5}$	$7.30x10^{5}$	$8.26x10^{5}$	$6.55x10^{5}$	$19.2x10^{5}$
curies/year	$.024x10^{9}$	$1.36x10^{9}$	$2.45x10^{9}$	$2.77x10^{9}$	$2.19x10^{9}$	$6.45x10^{9}$
watts/year	$.141x10^{4}$	$8.05x10^{4}$	$14.5x10^{4}$	$16.4x10^{4}$	$13.0x10^{4}$	$38.2x10^{4}$

Fast-Breeder Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year			$1.37x10^{5}$	$8.26x10^{5}$	$35.3x10^{5}$	$106X10^{5}$
curies/year			$0.46x10^{9}$	$2.77x10^{9}$	$11.8x10^{9}$	$35.6x10^{9}$
watts/year			$2.73x10^{4}$	$16.5x10^{4}$	$70.2x10^{4}$	$211x10^{4}$

TABLE 2

CERIUM-144 CONTENT IN HIGH-LEVEL WASTE PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production	Calendar Year					
	1970	1980	1985	1990	2000	2020
grams/year	$.15\times10^{5}$	8.26×10^{5}	14.9×10^{5}	16.8×10^{5}	13.3×10^{5}	39.2×10^{5}
curies/year	$.046\times10^{9}$	2.64×10^{9}	4.75×10^{9}	5.38×10^{9}	4.26×10^{9}	12.5×10^{9}
watts/year	$.041\times10^{6}$	2.31×10^{6}	4.17×10^{6}	4.72×10^{6}	3.74×10^{6}	11.0×10^{6}

Fast-Breeder Reactor Fuels

Annual Production	Calendar Year					
	1970	1980	1985	1990	2000	2020
grams/year			1.43×10^{5}	8.6×10^{5}	36.7×10^{5}	110×10^{5}
curies/year			$.46\times10^{9}$	2.75×10^{9}	11.8×10^{9}	35.3×10^{9}
watts/year			$.40\times10^{6}$	2.41×10^{6}	10.3×10^{6}	30.9×10^{6}

TABLE 3

PROMETHIUM-147 CONTENT IN HIGH-LEVEL WASTE PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year	$.058 \times 10^{5}$	3.32×10^{5}	5.97×10^{5}	6.75×10^{5}	5.35×10^{5}	15.7×10^{5}
curies/year	$.054 \times 10^{8}$	3.08×10^{8}	5.54×10^{8}	6.27×10^{8}	4.97×10^{8}	14.6×10^{8}
watts/year	$.028 \times 10^{5}$	1.58×10^{5}	2.85×10^{5}	3.23×10^{5}	2.56×10^{5}	7.52×10^{5}

Fast-Breeder Reactor Fuels

Annual Production	Calendar Year 1970	1980	1985	1990	2000	2020
grams/year			1.36×10^{5}	8.17×10^{5}	34.9×10^{5}	$10^{5} \times 10^{5}$
curies/year			1.26×10^{8}	7.59×10^{8}	32.4×10^{8}	97.4×10^{8}
watts/year			$.65 \times 10^{5}$	3.91×10^{5}	16.7×10^{5}	50.2×10^{5}

Sources and Types of Wastes

Ruthenium-106, cerium-144, and promethium-147 are only a few of the many fission products produced in nuclear reactors and are contained within the spent fuel elements. The spent fuel elements are removed from the reactor and processed for the recovery of usable fissionable materials. These radionuclides and the other high-level wastes are contained in the aqueous effluent from the spent-fuel processing step. The range of chemical compositions of these waste streams have been tabulated.[0715] These waste streams are primarily aqueous solutions of inorganic nitrate salts and any differences in their composition occurs mainly in the amounts and types of salts added during the processing step. The amount and activity of Ru-106, Ce-144, and Pm-147 present in the high-level waste streams after 1 year and 10 years is attached (Table 4).

These radionuclides can also be found in the secondary waste streams generated at the spent fuel processing plant. The volume of these waste streams can be quite large but the radionuclide activity in these streams is quite low. Ruthenium-106 which usually oxidizes and volatilizes during the fuel reprocessing step is also found in the off-gas streams.

Physical and Chemical Properties

The physical and chemical properties of ruthenium-106, cerium-144, and promethium-147 are included in the attached worksheets. Ruthenium-106 exhibits all possible positive valences; consequently, its chemistry is exceedingly complex. In the lower oxidation states it is basic in nature, while in the higher oxidation states it tends to be acidic. In alkaline solutions it may be oxidized to the ruthenite, ruthenate, perruthenate, or the tetroxide, depending upon the strength of the oxidizing agent. The tetroxide is volatile, boiling at around 100 C.

Cerium-144 is a member of the rare-earth group. Cerium has a valence of +3 but can also assume a valence of +4. Tetravalent cerium is a powerful oxidizing agent. The hydroxides and nitrates of cerium are slightly soluble in water. The carbonates, oxalates, and phosphates

TABLE 4

RUTHENIUM-106, CERIUM-144, AND PROMETHIUM-147 CONTENT IN HIGH-LEVEL FUEL REPROCESSING WASTE[0705]

Light-Water Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Fission Products	After 1 year			After 10 years		
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
$Ru^{106}-Rh^{106}$	81	2,656	546,000	0.16	5	1,100
$Ce^{144}-Pr^{144}$	143	3,800	912,000	0.05	1.5	300
Pm^{147}	92	44	85,000	8	4	8,000
Other Fission Products	34,784	3,500	679,000	35,002	1,019	307,600

Fast Breeder Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 58 MW/MTU)

Fission Products	After 1 year			After 10 years		
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
$Ru^{106}-Rh^{106}$	204	6,660	1,370,000	0.41	14	2,760
$Ce^{144}-Pr^{144}$	177	4,700	1,128,000	0.06	2	370
Pm^{147}	300	150	279,000	28	13	26,000
Other Fission Products	34,219	2,290	653,000	34,872	737	251,870

are not soluble. Promethium-147 is also a member of the rare-earth group and its chemistry is similar to that of cerium.

2. RADIATION HAZARD

Ruthenium-106 and cerium-144 are dangerous radioactive materials. Promethium-147 is a moderately dangerous radioactive material. The effects of their radiation exposure are primarily dependent on the amount of radiation and the portion of the body affected. The effects of acute whole-body gamma radiation exposure are: (1) 5 to 25 rads, minimal dose detectable by chromosome analysis or other specialized analyses, but not by hemogram; (2) 50 to 75 rads, minimal acute dose readily detectable in a specific individual (e.g., one who presents himself as a possible exposure case); (3) 75 to 125 rads, minimal acute dose likely to produce vomiting in about 10 percent of people so exposed; (4) 150 to 200 rads, acute dose likely to produce transient disability and clear hematological changes in a majority of people so exposed; (5) 300 rads, median lethal dose for single short exposure.[2666] These effects are for a single large dose of radiation or a series of substantial doses in a short interval of time to the total body. Standards for prolonged exposure over a fifty-year period have defined the single dose limit in terms of maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter for a radiation worker and the accumulated dose limit is 5(N - 18), where N is the individual's age in years. Limits for the thyroid, bone, and other organs have also been defined.[0563]

Values of the total body burden for each radionuclide required to produce the maximum permissible dose rates defined above have been compiled.[0563] For ruthenium-106 the critical organ is the kidney and the maximum permissible body burden is 3 microcuries. For cerium-144 and promethium-147 the critical organ is the bone and the maximum permissible body burden is 5 microcuries and 60 microcuries, respectively.

Their radiological toxicity can also be expressed in terms of the dose delivered to a particular body organ following the inhalation of 1 microcurie. For each of these radionuclides the single inhalation of 1 microcurie will produce the following doses:

Isotope	Form	Organ	Dose
Ru-106	Insoluble	Lung	1.2 rem
Ru-106	Soluble	Kidney	0.04 rem
Ce-144	Insoluble	Lung	1.0 rem
Ce-144	Soluble	Bone	1.1 rem
Pm-147	Insoluble	Lung	0.07 rem
Pm-147	Soluble	Bone	0.2 rem

3. OTHER HAZARDS

Cerium-144 resembles aluminum in its pharmacological action as well as in its chemical properties. Besides its radiation hazard, cerium-144 is not toxic to the skin or mucous membranes by repeated exposure and is only slightly toxic if absorbed into the body by inhalation or injection.[0766] Its fire and explosive hazard is moderate. It ignites spontaneously in air at temperatures of 150 C to 180 C. The toxicity of cerium compounds is the same as cerium, except when the anion has a toxicity of its own.

The details of ruthenium-106 toxicity are unknown, but it is believed to be toxic.[0776] It is dangerous when heated to decomposition since it emits toxic fumes of ruthenium oxide. Data on the toxicity of promethium-147 were not available.

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

Since all three radionuclides are hazardous to man by inhalation, ingestion, or direct radiation exposure, care is exercised in their handling. Special procedures and radiation shielding are utilized in their handling. The beta particles emitted by Ru-106, Ce-144, and Pm-147 are of low energy and cannot penetrate more than 0.02 in. of water. The daughter products of Ru-106 and Ce-144 emit high energy beta particles which can penetrate up to 0.8 in. of water. The principal shielding problem associated with their daughter's beta radiation is the bremsstrahlung effect which results from the presence of highly penetrating rays resulting from the deflection of the beta particles by the nuclei in the shielding medium. Since the bremsstrahlung effect increases with atomic number, shielding materials of high atomic weight are not satisfactory. Ru-106, Ce-144, and their daughters also emit gamma radiation. The gamma radiation is highly penetrating and high-density shields, such as lead, are required to stop the radiation. To detect and control personnel exposure to their radiation all persons working with this material should wear dosimetry devices which directly indicate the dose. Commonly used devices are the film badge and the thermoluminescent dosimeters (TLD).

These materials are stored in controlled reservations in specially constructed containers which are protected by both a primary and a secondary containment barrier. Special monitoring systems and proper warning signs are located in the general area of the storage facility.

Ruthenium-106 and cerium-144 are classified as a transport group III radionuclide and promethium-147 as a transport group IV radionuclide by the Department of Transportation. The rules and regulations governing their transportation are given in the Code of Federal Regulations (CFR) Title 49--Transportation, Parts 170 to 190.[2150]

The Ru-106 and Ce-144 content is limited to 3 curies for a Type A package and 200 curies for a Type B package defined in 49CFR173. The Pm-147 content is limited to 20 curies for a Type A package and 200 curies for a Type B package. These limits for each radionuclide are increased to 20 and 5,000 curies if their physical form meets the requirements of a special form material. Their release rate is limited to zero under the specified accident conditions for Type A and B quantities.[2150] The allowable release of radioactivity from packages containing large quantities of these materials is limited to gases and contaminated coolant containing total radioactivity exceeding neither 0.1 percent of the total radioactivity of the package nor 10 curies under the hypothetical accident conditions prescribed in 49CRF173.

Disposal/Reuse

The disposal of these materials is governed by the AEC Manual Chapter 0524[0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The radiation protection standards for these two groups are attached (Table 5). These standards also define their maximum concentrations in air and water for exposure to either an individual or to the general population. For an individual, the safe release of these materials to the environment in an unrestricted area should not exceed the concentrations in microcuries per milliliter (μc/ml) listed below[2149]:

Isotope	Form	Concentration in Air	Concentration in Water
Ru-106	Soluble	$3x10^{-9}$ μc/ml	$1x10^{-5}$ μc/ml
Ru-106	Insoluble	$2x10^{-10}$ μc/ml	$1x10^{-5}$ μc/ml
Ce-144	Soluble	$3x10^{-10}$ μc/ml	$1x10^{-5}$ μc/ml
Ce-144	Insoluble	$2x10^{-10}$ μc/ml	$1x10^{-5}$ μc/ml
Pm-147	Soluble	$2x10^{-9}$ μc/ml	$2x10^{-4}$ μc/ml
Pm-147	Insoluble	$3x10^{-9}$ μc/ml	$2x10^{-4}$ μc/ml

TABLE 5

RADIATION PROTECTION STANDARDS FOR INDIVIDUALS AND POPULATION GROUPS FOR EXTERNAL AND INTERNAL EXPOSURE[2149]

Type of Exposure	Dose to Individuals at Points of Maximum Probably Exposure (rem per year)	Average Dose to a Suitable Population Sample (rem per year)
Whole body, gonads, or bone marrow	0.5	0.17
Thyroid or bone	1.5	0.5
Bone (alternate standards)	Body burden at 0.003 micrograms of radium 226 or its biological equivalent	Body burden of 0.001 micrograms of radium 226 or its biological equivalent

The concentrations for the safe release of these materials to the general population are one-third the above values.

The disposal by release into a sanitary sewage system is limited to 10 microcuries for Ru-106 and Ce-144 and to 100 microcuries for Pm-147. The disposal by burial in the soil at any one location and time is limited to 1,000 microcuries for Ru-106 and Ce-144 and to 10,000 microcuries for Pm-147.[2149]

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Recovery

Option No. 1 - Volatilization-Absorption. Ruthenium-106 removal from solutions can be accomplished by oxidizing the ruthenium to its tetroxide state and volatilizing it by simultaneous heating and gas sparging. In the tetroxide state, ruthenium boils at 100 C. Ruthenium can be removed from uranium nitrate solutions by sparging with air containing ozone. Ruthenium decontamination factors (ratio of initial to final concentration) of about 100 were obtained during an 8-hour sparge with 1 percent ozone in air.[0714] Ruthenium can also be oxidized to the volatile form by hot concentrated nitric acid. The volatized ruthenium is then recovered by absorption. Two such absorbents are pyrolusite and activated carbon. Decontamination factors with pyrolusite alone are less than 100.[0714] Activated carbon used subsequent to pyrolusite absorption increases the decontamination factor 10 times.

Option No. 2 - Scavenging-Precipitation Foam Separation. The removal of radionuclides from low-level radioactive waste water by scavenging-precipitation form separation has been studied by Oak Ridge National Laboratory.[0703] The process consists of two steps: (1) precipitating in a sludge-blanket clarification step, and (2) achieving final decontamination in a foam-separation column. Processing rates of 300 gal. per hour (gph) for scavenging-precipitation and 120 gph for foam separation have been achieved. Cerium-144 decontamination factors of greater than 20 have been obtained.[0703] Ruthenium-106 decontamination factors of only four were obtained. The low decontamination factor is expected since ruthenium is one of the most difficult radionuclides to remove by scavenging.

Option No. 3 - Scavenging-Precipitation Ion Exchange. In this process final decontamination is obtained by ion exchange columns from the scavenging-precipation process. The process includes a provision for the recycle of the ion exchange waste to the scavenging-precipitation step. All the removed radionuclides are concentrated in the clarifier sludge. For cerium-144 and promethium-147 the overall decontamination factors varied from 20 to 700 and for ruthenium-106 the overall decontamination factor varied from 1.5 to 8.[0705] For cerium-144 and promethium-147, the use of ion exchange resins with a scavenging-precipitation step is a fairly simple and efficient means of removing these radionuclides from low-level, radioactive aqueous wastes. For ruthenium-106 wastes this method is not satisfactory since ruthenium in the liquid form is not very susceptible to ion exchange.

Option No. 4 - Water Recycle. The water recycle process is used for decontaminating radioactive waste water and recycling the purified water for reuse. This process has been demonstrated at the pilot plant scale by Oak Ridge National Laboratory.[0707] The steps in the process include: (1) clarification by controlled addition of coagulants, (2) demineralization by cation-anion exchange, and (3) sorption on granular activated carbon. For optimum removal of the ruthenium-106 during the coagulation-clarification step the pH of the waste must range between 7 and 8 for proper alum floc formation. High decontamination factors for ruthenium-106 can only be obtained during the final step by sorption on activated carbon. The overall decontamination factor for ruthenium-106 was 1,230 and for cerium-144 it was 800. The method is an improvement to the treat-and-discharge methods, but further work is required until full-scale production use is obtained.

Disposal

Option No. 1 - Land Burial. Land burial of Ru-106, Ce-144 and Pm-147 wastes, in small concentrations, at approved sites that are acceptable from a geologic and hydrologic standpoint, is an acceptable means of disposal. Their concentrations should not be in excess of 10^4 times the maximum permissible concentration for the general population in 10CFR20.[2149] All wastes to be disposed of should be in a solid form and encapsulated in a suitable container. Liquid wastes should be solidified, preferably using asphalt, in accordance with the methods described in the radioactive waste solidification report. The burial trenches should be designed not to intercept the ground water table and constructed with a bottom drain and sump for water monitoring. The trenches should be covered with either asphalt or vegetation to limit infiltration of water. The burial site design, geology, and hydrology should be in conformance with the criteria used in selecting and licensing the present commercial burial sites.[1423] Since land burial is successfully practiced on a commercial scale and since these isotopes have moderate half-lives, it should be considered as the most satisfactory method of disposing of all dilute concentrations of these wastes.

Option No. 2 - Near-Surface Liquid Storage. Near-surface storage of aqueous wastes in stainless steel tanks encased in concrete and buried underground is not considered as a satisfactory means of disposal. Aqueous wastes have been stored in this manner over the past 25 years. The tanks range in size from 0.33 to 1.3 million gal. and are generally equipped with devices for measuring temperatures, liquid levels, and leaks. At the present time these tanks are considered as an interim storage technique due to a general lack of confidence in their long-term integrity.[0705] Since present regulations require the solidification of all reactor wastes within 5 years following reprocessing,[2149] the near-surface storage of these wastes in steel tanks should only be considered as a near-term storage technique and not as a permanent storage or disposal technique.

Option No. 3 - Near-Surface Solid Storage. The storage of high-level solidified Ru-106, Ce-144, and Pm-147 wastes in engineered surface facilities offers the best immediate method for storage of these wastes. The technology for these facilities has been developed. The wastes will be under surveillance and control and can be retrieved, should this be required. The aqueous wastes should be solidified and packaged in a suitable container (steel). Of the four high-level solidification processes developed, spray and phosphate glass solidification offer the best solidified waste characteristics (see Radioactive Waste Solidification report). The wastes will be stored in stainless steel-lined concrete vaults which will be either air or water cooled. The periodic replacement of waste containers probably will not be required since their activity is reduced by a factor of 1,000 in 10 years for Ru-106 wastes, in 21 years for Ce-144 wastes, and in 26 years for Pm-147 wastes. The replacement of the waste containers might be required if other long-lived isotopes are also present.

Option No. 4 - Salt Deposits. This method offers the best potential for the disposal of wastes from fuel reprocessing since bedded salt deposits are completely free of circulating ground waters. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive wastes is satisfactory.[0733] Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as an ultimate method of disposal. The reactor-produced wastes must be solidified and disposed of in the solid form encapsulated in a suitable container. The preferred solidification process is spray solidification. The solidified wastes are then buried in rooms carved in the salt deposits approximately 1,000 ft below the ground. The salt is a good heat transmitter, provides about the same radioactive shielding as concrete, and can heal its own fractures by plastic flow. Salt deposits should be considered for the disposal of these wastes at approved sites that are acceptable from a design and geological standpoint.

Option No. 5 - Bedrock Disposal. The disposal of liquid wastes along with other spent fuel processing wastes in vaults excavated in crystalline rock over 1,500 ft beneath the ground is currently being evaluated by E. I. du Pont de Nemours, at their Savannah River Plant near Aiken, South Carolina.[0894,1396] The wastes would be stored in six tunnels and once in the tunnels the wastes will seep into the surrounding rock. Located above the crystalline rock is the Tuscaloosa formation, a good source of freshwater, which is separated by a layer of clay that would act as a barrier to the leakage of radioactive wastes. An advisory committee appointed by the National Academy of Sciences recommended abandonment of the project. In May 1972, another National Academy of Sciences panel concluded that bedrock storage provides a reasonable prospect for long-term safe storage but precise information is needed to decide if and where underground storage vaults should be built. Another method has also been proposed for disposing of liquid wastes by in situ incorporation in molten silicate rock.[2146] At the present time both of these methods are unproved since sufficient engineering data or exploration has not been completed to verify their suitability.

Option No. 6 - Hydraulic Fracturing. The direct disposal of aqueous low-level radioactive wastes into shale formations has been investigated by Oak Ridge National Laboratory.[0709] The method consists of mixing the aqueous wastes with cement and pumping the resulting slurry down a well and out into a nearly horizontal fracture in a thick shale formation. Additional work is required to demonstrate that this method of disposal is satisfactory.

To summarize, ruthenium-106, cerium-144, and promethium-147 are radioactive isotopes that account for a majority of the initial activity present in spent fuel processing wastes. The acceptable method of treatment is solidification and interim storage in near-surface engineered facilities followed by permanent disposal in salt deposits. Cerium-144 and promethium-147 can be recovered from low-level aqueous waste streams by scavenging-precipitation ion exchange. Ruthenium-106 can be recovered by volatization and absorption on pyrolusite and activated carbon. Generally, during the reprocessing of uranium and plutonium it is desirable to prevent the volatization of ruthenium. This can be accomplished during the leaching operation by adding a reducing agent such as sodium nitrite or nitrogen dioxide to suppress the ruthenium volatization.

6. APPLICABILITY TO NATIONAL DISPOSAL SITES

Ruthenium-106, cerium-144, and promethium-147 are candidates for a National Disposal Site. From a cost and safety viewpoint, it would be desirable to combine the reprocessing plant and the National Disposal Site. The recommended treatment for the high-level wastes is solidification by either the spray or phosphate glass solidification processes, interim storage in near-surface, engineered facilities, and ultimate disposal in salt deposits.

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HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Ruthenium-106

Half-life 368 days

Type of Decay Negative Beta

Structural Formula

$_{44}Ru^{106}$

Molecular Wt. 106 Melting Pt. 2250 C Boiling Pt. 3900 C

Density 12.30 gm/cm^3 Specific Power 33.1 watts/gm

Solubility Specific Activity 3,360 curies/gm

Cold Water Insoluble Hot Water Insoluble

Others: Insoluble in aqua regia, acid & alcohol; soluble in fused alkali

Decay Chain

$$\underset{(368d)}{Ru^{106}} \xrightarrow{B^-} \underset{(30s)}{Rh^{106}} \xrightarrow{B^-} \underset{(stable)}{Pd^{106}}$$

Radiation Energy Level & Intensities

Ruthenium-106

Beta: 0.0392 Mev (max)
Gamma: None

Rhodium-106

Beta: 3.55 Mev (90%)
3.10 Mev (3%)
2.40 Mev (5%)
Gamma: 0.512 Mev (21%)
0.622 Mev (1%)
1.050 Mev (1.5%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Cerium-144

Half-life 284 days

Type of Decay Negative Beta

Structural Formula

$_{58}Ce^{144}$

Molecular Wt. 144 Melting Pt. 795 C Boiling Pt. 3468 C

Density 6.78 gm/cc Specific Power 25.6 watts/gm

Specific Activity 3,180 curies/gm

Solubility

Cold Water Slightly reactive Hot Water Reacts

Others: Soluble in dilute mineral acid; insoluble in alkali

Decay Chain

$$\underset{(284d)}{Ce^{144}} \xrightarrow{B^-} \underset{(17.3m)}{Pr^{144}} \xrightarrow{B^-} \underset{(stable)}{Nd^{144}}$$

Radiation Energy Level & Intensities

Cerium-144

Beta:	0.309	Mev	(76%)
	0.175	Mev	(24%)
Gamma:	0.134	Mev	(11%)
	0.080	Mev	(2%)

Praseodymium-144

Beta:	2.996	Mev	(max)
Gamma:	2.186	Mev	(0.7%)
	1.487	Mev	(0.3%)
	0.695	Mev	(1.5%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References:
(1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Promethium-147

Half-life 2.62 years

Type of Decay Negative beta

Molecular Wt. 147

Density 7.3 gm/cm^3

Structural Formula

$_{61}Pm^{147}$

Melting Pt. 1035 C

Boiling Pt. 2730 C

Specific Power 0.33 watts/gm

Specific Activity 929 curies/gm

Solubility

Cold Water

Hot Water

Others:

Decay Chain

$Pm^{147} \xrightarrow{B^-} Sm^{147}$

(2.62y) (stable)

Radiation Energy Level & Intensities

Beta: 0.224 Mev (100%)

Gamma: None

Shipping Regulations Classified as a transport group IV radionuclide by the Department of Transportation

Comments Very limited chemical and physical data. Is a member of the rare-earth group.

References: (1) 2147
(2) 2150

PROFILE REPORT

Strontium-90 (Yttrium-90)

1. GENERAL

Introduction

Strontium-90 is a radioactive isotope principally produced by the fission of uranium and plutonium. It has a half-life of 28 years and emits a 0.546 Mev beta particle. Strontium-90 produces a short half-lived daughter, yttrium-90. It has a half-life of 64 hours and emits a 2.27 Mev beta particle. Yttrium-90, in turn, produces zirconium-90 which is a stable element.

Strontium-90 has attracted great interest as a public health hazard since it is the most biologically significant of the radioactive fission products produced in either nuclear weapon tests or nuclear reactors. Its biological significance is derived from several factors: (1) it has a large fission yield (5.9%); (2) it has a long effective half-life; and (3) it tends to deposit and concentrate in the bone tissue (due to the fact that strontium is chemically similar to calcium). In the wastes generated at nuclear power plants strontium-90 and its daughter yttrium-90 are responsible for approximately 7 percent and 38 percent of the total fission product activity in the wastes after 1 year and 10 years, respectively.

The projected growth in the production of strontium-90 will parallel that of the civilian nuclear power program. Present projections indicate that nuclear power will account for 30 percent of the total power production by the year 1980.[2148] Projected annual production figures from the civilian nuclear power program for strontium-90 to the year 2020 for light-water and fast-breeder reactor fuels are attached (Table 1). The expected total accumulated radioactivity for strontium-90 from the year 1970 to 2020 in millions of curies is:

Year	Radioactivity (Megacuries)
1970	4
1980	962
1990	4,640
2000	9,550
2020	29,400

These figures along with the annual production figures illustrate the tremendous growth anticipated in the production of this isotope. Annual production figures or accumulated totals for strontium-90 produced as part of the nuclear weapons program were not available.

Manufacture

Strontium-90 is produced by the fission of the heavy elements, i.e., those elements heavier than lead. The fission reaction results from neutron bombardment or spontaneous decomposition of the nucleus. A typical fission reaction is:

$${}_{92}U^{235} + {}_{o}n^{1} \longrightarrow {}_{38}Sr^{90} + {}_{54}Xe^{144} + 2{}_{o}n^{1}$$

Presently, strontium-90 is produced in thermal power reactors by the fission of uranium. Civilian nuclear power plants are the major producers and a smaller amount is produced at AEC facilities. At the present time there are 22 nuclear power plants in operation with an additional 104 being built or planned.[2151] In the future, strontium-90 will also be produced in fast-breeder reactors by the fission of plutonium.

TABLE 1

STRONTIUM-90 CONTENT IN HIGH-LEVEL WASTE PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production	Calendar Year					
	1970	1980	1985	1990	2000	2020
grams/year	2.83×10^4	161×10^4	291×10^4	328×10^4	260×10^4	764×10^4
curies/year	4.0×10^6	228×10^6	410×10^6	463×10^6	367×10^6	1080×10^6
watts/year	$.52\times10^4$	29.6×10^4	53.3×10^4	60.30×10^4	47.8×10^4	140.5×10^4

Fast-Breeder Reactor Fuels

Annual Production	Calendar Year			
	1985	1990	2000	2020
grams/year	11×10^4	66×10^4	282×10^4	846×10^4
curies/year	15.5×10^6	93.4×10^6	399×10^6	1200×10^6
watts/year	2.0×10^4	12.2×10^4	52×10^4	156×10^4

Uses

Strontium-90 is a long-lived high-energy beta emitter. It is used in military and space applications as an energy source for small auxiliary power units. It is also used in the radioactive gauge field. Beta gauges activated by strontium-90 are extensively used in industry to measure and control the thickness or density of thin materials (paper, textiles, etc.) or coatings. In the field of nuclear medicine strontium-90 is used as a source of beta radiation for the treatment of skin diseases and eye disorders and also as a medical blood irradiator. At the present time the strontium-90 used in the above applications is obtained from the waste streams in the Richland, Washington, chemical separations plant and is distributed through the Oak Ridge National Laboratory, Isotopes Sales Department. In the future, when its production is warranted by market demands, it could be available in large quantities from commercial firms operating chemical processing plants. The AEC revenue from the distribution of strontium-90 through the fiscal years 1965 to 1971 is[215]:

Year	Revenue (Dollars)
1965	3,000
1966	65,000
1967	59,000
1968	188,000
1969	169,000
1970	72,000
1971	248,000

These figures illustrate the relatively small commercial market for the use of strontium-90 at the present.

Sources and Types of Strontium-90 Wastes

Although produced in nuclear power reactors the primary source of strontium-90 is in the high-level aqueous waste streams generated at the spent fuel processing plants. The range of chemical compositions of the various types of waste streams obtained from the processing step have been tabulated,[0715] and in all cases are primarily aqueous solutions of inorganic nitrate salts. Strontium-90 is normally found in the waste stream in the form of a nitrate or oxide. The characteristics of strontium-90 products in the high-level wastes at two different time periods is attached (Table 2).

Strontium-90 is also found in the low-level waste streams generated at spent fuel processing facilities. The strontium-90 activity in these waste streams is from a few hundredths to several tenths of a curie per gallon of waste. Strontium-90 is also found in the wastes resulting from research laboratories and medical and industrial applications. Generally the strontium-90 waste from these sources is found in the solid form.

Physical and Chemical Properties

The physical and chemical properties of strontium-90 and its short half-lived daughter yttrium-90 are included in the attached worksheet. Strontium-90 is a member of the alkaline-earth metal group. It is a highly metallic and highly electropositive element with a valence of +2. It dissolves readily in acids and will burn when heated in air, oxygen, or carbon dioxide. At low temperatures, oxidation is slow owing to the formation of a protective oxide film. Strontium-90 reacts readily with water, releasing hydrogen and forming metal and hydroxyl ions. The carbonates and sulfates of strontium are very insoluble. The fluorides and the oxalate of strontium are also insoluble. The sulfides, however, are soluble in water.

2. RADIATION HAZARD

Strontium-90 is a very dangerous radioactive material. It tends to concentrate in the bone and irradiate the adjacent soft tissue. The symptoms of strontium-90 radiation exposure are nausea and fatigue followed by vomiting and diarrhea. The possible types of injury include:

TABLE 2
STRONTIUM-90 CONTENT IN HIGH-LEVEL FUEL REPROCESSING WASTES[0705]

Light-Water Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Fission Products	After 1 year Grams/Tonne	Watts /Tonne	Curies/Tonne	After 10 years Grams/Tonne	Watts /Tonne	Curies/Tonne
Sr^{90} - Y^{90}	534	530	151,000	427	425	120,900
Other Fission Products	34,566	9,470	2,071,000	34,673	605	196,100

Fast-Breeder Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 58 MW/MTU)

Fission Products	After 1 year Grams/Tonne	Watts/Tonne	Curies/Tonne	After 10 years Grams/Tonne	Watts/Tonne	Curies/Tonne
Sr^{90} - Y^{90}	300	298	85,000	241	239	68,000
Other Fission Products	34,600	13,502	3,345,000	34,658	529	213,000

leukopenia, anemia, leukemia, cataracts, and increases in the average rate of genetic mutation. The effects of strontium-90 radiation exposure are primarily dependent on the amount of radiation and the portion of the body affected. Standards for prolonged exposure over a 50-year period have defined the single dose limit in terms of the maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter for a radiation worker and the accumulated dose limit is 5(N - 18), where N is the individual's age in years. Limits for the thyroid, bone, and other organs have also been defined (Table 3).[0563]

Following the inhalation of 1 microcurie of strontium-90 in the insoluble form the critical organ is the bone and the dose delivered to the bone following the inhalation of 1 microcurie of strontium-90 is 56 rem. The dose delivered to the bone following injection of 1 microcurie into the body via a wound is 90 rem. Values of the maximum permissible total body burden of strontium-90 which are deposited in the total body and produces the maximum permissible dose rate to a particular body organ have been compiled.[0563] The maximum permissible body burden for the bone is 2.0 microcuries and for the total body 20 microcuries. The above body burden rates can be expressed in terms of the maximum permissible concentrations of strontium-90 in air and water. These concentrations can be expressed in terms of a daily intake of air and water at the continuous exposure rate of 168 hours per week for a period of 50 years. For strontium-90 the critical organ is the bone and the maximum permissible concentration in water is $1\text{x}10^{-6}$ microcuries per milliliter and in air $1\text{x}10^{-10}$ microcuries per milliliter.[0563]

3. OTHER HAZARDS

Strontium-90 resembles calcium in its metabolism and behavior. Strontium-90, besides its radiation hazard, is moderately toxic to the skin or mucous membranes following a single exposure. Its fire and explosive hazard is moderate, in the form of dust, when exposed to flame or chemical reaction.[0766] It reacts with water to evolve hydrogen. Strontium nitrate or peroxide are powerful oxiding materials. In

TABLE 3

RADIATION PROTECTION STANDARDS FOR INDIVIDUALS AND POPULATION GROUPS FOR EXTERNAL AND INTERNAL EXPOSURE[2149]

Type of Exposure	Dose to Individuals at Points of Maximum Probable Exposure (rem per year)	Average Dose to a Suitable Population Sample (rem per year)
Whole body, gonads, or bone marrow	0.5	0.17
Thyroid or bone	1.5	0.5
Bone (alternate standards)	Body burden at 0.003 micrograms of radium 226 or its biological equivalent	Body burden of 0.001 micrograms of radium 226 or its biological equivalent

contact with easily oxidizable substances they react rapidly to cause ignition or explosion by friction or on contact with a small amount of water.

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

Since strontium-90 is hazardous to man by inhalation, ingestion, or direct radiation exposure, care is exercised in its handling. Special procedures and radiation shielding are required in the handling of strontium-90. The beta particles emitted by strontium-90 generally cannot penetrate more than one centimeter of metal or glass. The principal problem associated with the beta radiation is the bremsstrahlung effect which results from the presence of highly penetrating rays resulting from the deflection of beta particles by the nuclei in the shielding medium. Since the bremsstrahlung effect increases with atomic number, shielding materials of high atomic weight are not satisfactory. Lead and high-density concrete are commonly used shielding materials for strontium-90 radiation. To detect and control personnel exposure to strontium-90 radiation all persons working with this material should wear dosimetry devices which directly indicate the dose. Commonly used devices are the film badge and the thermoluminescent dosimeters (TLD).

Strontium-90 is stored in controlled reservations in specially constructed containers. The strontium-90 is protected by both a primary and a secondary containment barrier. Special monitoring systems and proper warning signs are located in the general area of the storage facility.

Strontium-90 is classified as a transport group II radionuclide by the Department of Transportation, and the rules and regulations governing their transportation are given in the Code of Federal Regulations (CFR) Title 49--Transportation, Parts 170 to 190.[2150] The strontium-90 content is limited to 0.05 curies for a Type A package and 20 curies for a Type B package defined in 49CFR173. The limits are increased to 20 and 500 curies if the physical form of the strontium-90 meets the requirements of a special form material. The strontium-90 release rate is limited to zero under the specified accident conditions for Type A and B quantities.[2150] The allowable release of radioactivity from packages containing large quantities of strontium-90 is limited to gases and contaminated coolant containing total radioactivity exceeding neither 0.1 percent of the total radioactivity of the package nor 0.5 curies under the hypothetical accident conditions prescribed in 49CFR173.

Disposal/Reuse

The disposal of strontium-90 is governed by the AEC Manual Chapter 0524[0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The radiation protection standards for these two groups is attached. The standards for strontium-90 also define its maximum concentrations in air and water in microcuries per milliliter (μc/ml) listed below[2149]:

Exposure Group	Strontium-90 Form	Concentration in Air	Concentration in Water
Individual	Soluble	3×10^{-11} μc/ml	3×10^{-7} μc/ml
Individual	Insoluble	2×10^{-10} μc/ml	4×10^{-5} μc/ml
Population	Soluble	1×10^{-11} μc/ml	1×10^{-7} μc/ml
Population	Insoluble	$.67\times10^{-10}$ μc/ml	1.3×10^{-5} μc/ml

Although rarely utilized, the disposal of strontium-90 by release into a sanitary sewage system is limited to 1 microcurie or a concentration of 1×10^{-5} microcuries per milliliter of water in the soluble form or 1×10^{-3} microcuries per milliliter of water in the insoluble form. The disposal of strontium-90 by burial in the soil at any one location and time is limited to 100 microcuries of strontium-90.[2149]

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Recovery

Option No. 1 - Precipitation. Strontium-90 can be removed from high-level processing wastes following the spent fuel processing step by precipitating as a carbonate or fluoride[0686] or absorbing on alumino-silicate zeolites.[0738] The standard procedure for the recovery of strontium-90 is to precipitate as a carbonate and then add titanium dioxide and heat to drive off the CO_2, forming strontium titanite. The strontium-90 can be recovered from the other high-level wastes for commercial purposes, or to provide isolation of the strontium-90 for heat transfer and safety reasons (see Radioactive Waste Solidification Report).

Option No. 2 - Scavenging-Precipitation Foam Separation. The removal of strontium-90 from radioactive waste water by scavenging-precipitation foam separation has been studied by Oak Ridge National Laboratory.[0703] The process consists of two steps: (1) precipitating in a sludge blanket clarification step, and (2) achieving final decontamination in a foam-separation column. Processing rates of 300 gal. per hour (gph) for scavenging precipitation and 120 gph for foam separation have been achieved. Strontium-90 is precipitated by the addition of calcium carbonate or calcium phosphate. Strontium-90 decontamination factors (ratio of initial to final concentration) of 1050 have been obtained.[0703] At the present time, further development is required to reduce operating costs and increase processing rates.

Option No. 3 - Scavenging-Precipitation Ion Exchange. In this process final decontamination is obtained by ion exchange columns from the scavenging-precipitation process. In the phenolic ion exchange column calcium and magnesium are removed with the strontium. The process includes a provision for the recycle of the ion exchange waste to the scavenging-precipitation step. All the removed radionuclides are concentrated in the clarifier sludge. The overall decontamination factors for strontium-90 varied from 1,200 to 12,000.[0705] The use of ion exchange resins with a scavenging-precipitation step is a fairly simple and efficient means of removing strontium-90 from low-level radioactive aqueous wastes.

Option No. 4 - Water Recycle. The water recycle process is used for decontaminating radioactive waste water and recycling the purified water for reuse. This process has been demonstrated at the pilot plant scale by Oak Ridge National Laboratory.[0707] The steps in the process include: (1) clarification by controlled addition of coagulants; (2) demineralization by cation-anion exchange; and (3) sorption on granular activated carbon. The majority of the strontium-90 is removed by the cation-anion exchange. The strontium-90 overall decontamination factor was 5,700. The method is an improvement to the treat-and-discharge methods, but further work is required until full-scale production use is obtained.

Storage/Disposal

Option No. 1 - Land Burial. Land burial of strontium-90 wastes, in small concentrations, at approved sites that are acceptable from a geologic and hydrologic standpoint, is an acceptable means of disposal. Strontium-90 concentration should not be in excess of 10^4 times the maximum permissible concentration for the general population in 10CFR20.[2149] All strontium-90 wastes to be disposed of should be in a solid form and packaged in a suitable container. Liquid strontium-90 wastes should be solidified, preferably using asphalt, in accordance with the methods described in the radioactive waste solidification report. The burial trenches should be designed not to intercept the ground water table and constructed with a bottom drain and sump for water monitoring. The trenches should be covered with either asphalt or vegetation to limit infiltration of water. The burial site design, geology, and hydrology should be in conformance with the criteria used in selecting and licensing the present commercial burial sites.[1423] Since land burial has successfully been practiced on a commercial scale, it should be considered as the most satisfactory method of disposing of all dilute concentrations of strontium-90 wastes.

Option No. 2 - Near-Surface Liquid Storage. Near-surface storage of reactor-produced aqueous solutions of strontium-90 and other fission products salts which are stored in carbon steel or stainless steel tanks encased in concrete and buried underground is not considered as a satisfactory means of disposal. Aqueous solutions of high-level wastes have been stored in this manner over the past 25 years. The tanks range in size from 0.33 to 1.3 million gal. and are generally equipped with devices for measuring temperatures, liquid levels, and leaks. These tanks are considered as an interim storage technique due to a general lack of confidence in their long-term integrity.[0705] Since present regulations require the solidification of all reactor wastes within 5 years following reprocessing,[2149] the near-surface storage of aqueous solutions of strontium-90 and other fission product salts in steel tanks should only be considered as a near-term storage technique and not as a permanent storage or disposal technique.

Option No. 3 - Near-Surface Solid Storage. The storage of solidified, high-level wastes containing strontium-90 and other waste salts in engineered storage facilities offers the best intermediate method for storage of these wastes. The technology for these facilities has been developed. The wastes will be under surveillance and control and can be retrieved, should this be required. The high-level wastes from fuel reprocessing should be solidified and packaged in a suitable container (steel). Of the four high-level solidification processes developed, spray or phosphate glass solidification offer the best solidified waste characteristics (see Radioactive Waste Solidification report). The wastes will be stored in stainless steel-lined concrete vaults which will be either air or water cooled.

Option No. 4 - Salt Deposits. This method offers the best potential for the disposal of wastes from fuel reprocessing since bedded salt deposits are completely free of circulating ground waters. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November of 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive wastes is satisfactory.[0733] Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as an ultimate method of disposal. The wastes containing strontium-90 must be solidified and disposed of in the solid form and packaged in a suitable container. The solidified wastes are then buried in rooms carved in the salt deposits approximately 1,000 ft below the ground. The salt is a good heat transmitter, provides about the same radioactive shielding as concrete, and can heal its own fractures by plastic flow. The critical problem is the selection of a site that meets the necessary design and geological criteria for the mixture of fission product and actinide wastes.

Option No. 5 - Bedrock Disposal. The disposal of low-level aqueous solutions of strontium-90 wastes along with other spent fuel reprocessing wastes in vaults excavated in crystalline rock over 1,500 ft beneath the ground is currently being evaluated by E. I. du Pont de Nemours, at their Savannah River Plant near Aiken, South Carolina.[0894,1396] The wastes would be stored in six tunnels and once in the tunnels the wastes will seep into the surrounding rock. Located above the crystalline rock is the Tuscaloosa formation, a good source of freshwater, which is separated by a layer of clay that would act as a barrier to the leakage of radioactive wastes. An advisory committee appointed by the National Academy of Sciences recommended abandonment of the project. In May 1972, another National Academy of Sciences panel concluded that bedrock storage provides a reasonable prospect for long-term safe storage, but precise information is needed to decide if and where underground storage vaults should be built. Another method has also been proposed for disposing of liquid wastes by in situ incorporation in molten silicate rock.[2146] At the present time both of these methods are unproven since sufficient engineering data or exploration has not been completed to verify their suitability. Due to the long half-lives of certain of the elements of the mixed fission products and actinides and high biological hazard, it is doubtful that data could ever be accumulated to prove that the geological characteristics of the site over the next few hundred years are acceptable to ensure the absolute safety of such a disposal method.

Option No.6 - Hydraulic Fracturing. The direct disposal of aqueous, low-level radioactive wastes into shale formations has been investigated by Oak Ridge National Laboratory.[0709] The method consists of mixing the aqueous wastes with cement and pumping the resulting slurry down a well and out into a nearly horizontal fracture in a thick shale formation. Since strontium-90 is leached from cement and no additional encapsulation is provided and since it has long half-life this method of disposal is not recommended.

To summarize, strontium-90 can be recovered from the high-level waste streams from the spent fuel processing facilities for separate disposal or reuse. The separation of strontium-90 from these waste streams will probably be required with the introduction of the fast-breeder reactor due to its high heat content (see Radioactive Waste Solidification report). The acceptable method of treatment is spray or phosphate glass solidification, followed by storage in near-surface engineered facilities. Finally, these wastes should be disposed of in salt deposits. Strontium-90 should be recovered from the low-level waste streams to minimize the amount directly released to the environment. An adequate method of recovery is scavenging-precipitation ion exchange. The recovered strontium-90 can then be solidified, preferably using asphalt, and disposed of at approved sites by land burial.

6. APPLICABILITY TO NATIONAL DISPOSAL SITES

Strontium-90 is a candidate for a National Disposal Site due to its health hazard and its projected growth with that of the civilian nuclear power program. From a cost and safety viewpoint, it would be desirable to combine the reprocessing plant and the National Disposal Site. Interim storage in near-surface engineered facilities offers considerable latitude in the storage site selection. The site selection for ultimate disposal of these wastes is limited to particular geological areas in which salt deposits are present.

The recommended treatment for strontium-90 in low-level waste streams is recovery by scavenging-precipitation ion exchange followed by solidification with asphalt and disposal by land burial. For the high-level, strontium-90 wastes, the recommended processes are recovery by either spray or phosphate glass solidification, interim storage of the solidified waste in near-surface engineered storage facilities, and disposal in salt beds.

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HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Strontium-90

Half-life 28 years

Type of Decay Negative Beta

Structural Formula

$_{38}\mathrm{Sr}^{90}$

Molecular Wt. 90 Melting Pt. 769C Boiling Pt. 1384C

Density 2.6 gm/cc Specific Power 0.98 watts/gm

Specific Activity 142 curies/gm

Solubility

Cold Water Reacts Hot Water Reacts

Others: Soluble in acid, alcohol, and liquid NH_3

Decay Chain

$$\underset{(28y)}{\mathrm{Sr}^{90}} \xrightarrow{B^-} \underset{(64h)}{\mathrm{Y}^{90}}$$

Radiation Energy Level & Intensities

Beta: 0.549 Mev (100%)

Gamma: None

Shipping Regulations Classified as a transport group II radionuclide by the Department of Transportation

Comments

References:
(1) 0766
(2) 2147
(3) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Yttrium-90

Half-life 64 hours

Type of Decay Negative Beta

Structural Formula

$_{39}Y^{90}$

Molecular Wt. 90 Melting Pt. 1495C Boiling Pt. 2927C

Density 4.34 Specific Power

Solubility Specific Activity 5.3×10^5 curies/gm

Cold Water Slightly reactive Hot Water Reacts

Others: Very soluble in dilute acid; soluble in hot KOH

Decay Chain

$$\underset{(64h)}{Y^{90}} \xrightarrow{B^-} \underset{(Stable)}{Zr^{90}}$$

Radiation Energy Level & Intensities

Beta: 2.27 Mev(max)

Gamma: None

Shipping Regulations Classified as a transport group IV radionuclide by the Department of Transportation

Comments

References: (1) 0766
(2) 2150

PROFILE REPORT

Zirconium-95, Niobium-95

1. GENERAL

Zirconium-95 is a radioactive isotope principally produced in nuclear reactors by the fission of uranium and plutonium. Zirconium-95 has a half-life of 65 days and emits both beta particles and gamma rays. Zirconium-95 produces, by beta decay, niobium-95. Niobium-95 has a half-life of 35 days, emits a 0.160 Mev beta particle and a 0.765 Mev gamma ray. Niobium-95 is transformed by beta decay into molybdenum-95 which is a stable element.

Zirconium-95 and niobium-95 are both short-lived isotopes and account for 25 percent of the total activity in spent fuel processing wastes after 90 days. Their activity decreases to less than 1 percent of the total after 1 year. They also present considerable difficulty in the separation of uranium and plutonium from these elements since they both form radioactive colloids in solution. They also tend to be absorbed on surfaces such as container walls.

The projected growth in zirconium-95 and niobium-95 will parallel that of the civilian nuclear power program. Present projections indicate that nuclear power will account for 30 percent of the total power production by the year 1980.[2148] Projected annual production figures from the civilian nuclear power program for these two isotopes to the year 2020 for light-water and fast-breeder reactor fuels are attached (Tables 1 and 2). These figures illustrate the tremendous growth anticipated in their production.

TABLE 1

ZIRCONIUM-95 CONTENT IN HIGH-LEVEL WASTES PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production			Calendar Year			
	1970	1980	1985	1990	2000	2020
grams/year	$.0128\times10^4$	7.31×10^4	13.2×10^4	14.9×10^4	11.8×10^4	39.7×10^4
curies/year	$.272\times10^8$	15.5×10^8	27.9×10^8	31.6×10^8	25.0×10^8	73.6×10^8
watts/year	$.142\times10^6$	8.11×10^6	14.6×10^6	16.5×10^6	13.1×10^6	38.5×10^6

Fast-Breeder Reactor Fuels

Annual Production			Calendar Year			
	1970	1980	1985	1990	2000	2020
grams/year			3.54×10^4	21.3×10^4	91.1×10^4	274×10^4
curies/year			7.50×10^8	45.2×10^8	193×10^8	580×10^8
watts/year			3.93×10^6	23.7×10^6	101×10^6	304×10^6

TABLE 2

NIOBIUM-95 CONTENT IN HIGH-LEVEL WASTE PRODUCED BY THE CIVILIAN NUCLEAR POWER PROGRAM[0705]

Light-Water Reactor Fuels

Annual Production			Calendar Year			
	1970	1980	1985	1990	2000	2020
grams/year	$.012\times10^4$	6.54×10^4	11.8×10^4	13.3×10^4	10.6×10^4	31.1×10^4
curies/year	$.45\times10^8$	25.7×10^8	46.3×10^8	52.4×10^8	41.5×10^8	122×10^8
watts/year	$.22\times10^6$	12.3×10^6	22.2×10^6	25.1×10^6	19.9×10^6	58.6×10^6

Fast-Breeder Reactor Fuels

Annual Production			Calendar Year			
	1970	1980	1985	1990	2000	2020
grams/year			2.42×10^4	14.6×10^4	62.2×10^4	187×10^4
curies/year			9.50×10^8	57.2×10^8	244×10^8	734×10^8
watts/year			4.56×10^6	27.5×10^6	118×10^6	353×10^6

The primary sources of zirconium-95 and niobium-95 wastes are the spent fuel processing plants. These isotopes and the other resultant high-level radioactive wastes are contained in the aqueous effluent from the spent fuel processing step. The range of chemical compositions of the various types of waste streams obtained from the spent fuel processing step have been tabulated.[0715] The activity of zirconium-95 and niobium-95 in the spent reactor wastes at two different times is attached (Tables 3 and 4). Due to their short half-lives, their activity after 10 years is reduced to zero. They are also found in very small quantities in the low-level radioactive waste streams generated at spent fuel processing facilities.

The physical and chemical properties of zirconium-95 and niobium-95 are included in the attached worksheets. Zirconium-95 is a member of the group IV series of metals, all of which have a characteristic +4 oxidation state. Zirconium-95 is not readily soluble in nitric acid but dissolves at substantial rates in both hydrofluoric and moderately concentrated sulfuric acid. Niobium-95 exists in the +5 oxidation state in a majority of its compounds. Niobium-95 is a passive metal and does not dissolve in most acids when pure, but is attacked by these acids if impurities are present. Both isotopes tend to form radioactive colloids in solution which do not act as true solutes but tend to be absorbed on surfaces.

2. RADIATION HAZARD

Zirconium-95 and niobium-95 both are moderately dangerous radioactive materials. Zirconium-95 is classified as a slightly more hazardous material than niobium-95. The effects of their radiation are primarily dependent on the amount of radiation and the portion of the body affected. The effects of acute whole-body gamma radiation exposure are: (1) 5 to 25 rads, minimal dose detectable by chromosome analysis or other specialized analyses, but not by hemogram; (2) 50 to 75 rads, minimal acute dose readily detectable in a specific individual (e.g., one who presents himself as a possible exposure case);

TABLE 3

ZIRCONIUM-95 CONTENT IN HIGH-LEVEL FUEL REPROCESSING WASTES[0705]

Light-Water Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Fission Products	After 1 year Grams/Tonne	 Watts/Tonne	 Curies/Tonne	After 10 years Grams/Tonne	 Watts/Tonne	 Curies/Tonne
Zr^{95}	1.3	146	28,000	0	0	0
Other Fission Products	35,099	9,854	2,194,000	35,100	1030	317,000

Fast-Breeder Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 58 MW/MTU)

Fission Products	After 1 year Grams/Tonne	 Watts/Tonne	 Curies/Tonne	After 10 years Grams/Tonne	 Watts/Tonne	 Curies/Tonne
Zr^{95}	630	310	59,000	0	0	0
Other Fission Products	34,897	13,490	3,371,000	34,900	776	281,000

TABLE 4

NIOBIUM-95 CONTENT IN HIGH-LEVEL FUEL REPROCESSING WASTES[0705]

Light-Water Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 30 MW/MTU)

Fission Products		After 1 year			After 10 years	
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
Nb^{95}	2	285	59,000	0	0	0
Other Fission Products	35,098	9,715	2,163,000	35,100	1030	317,000

Fast-Breeder Reactor Fuels
(Fuel Exposed to 33,000 MWD/MTU at 58 MW/MTU)

Fission Products		After 1 year			After 10 years	
	Grams/Tonne	Watts/Tonne	Curies/Tonne	Grams/Tonne	Watts/Tonne	Curies/Tonne
Nb^{95}	3	600	125,000	0	0	0
Other Fission Products	34,897	13,200	3,305,000	34,900	776	281,000

(3) 75 to 125 rads, minimal acute dose likely to produce vomiting in about 10 percent of people so exposed; (4) 150 to 200 rads, acute dose likely to produce transient disability and clear hematological changes in a majority of people so exposed; (5) 300 rads, median lethal dose for single short exposure.[2666] These effects are for a single large dose of radiation or a series of substantial doses in a short interval of time to the total body. Standards for prolonged exposure over a 50-year period have defined the single dose limit in terms of the maximum permissible dose accumulated in a period of 13 weeks. The whole body exposure limit is 3 rem per quarter for a radiation worker and the accumulated dose limit is 5(N - 18), where N is the individual's age in years. Limits for the thyroid, bone, and other organs have been defined (Table 5).[0563]

The zirconium-95 dose in the insoluble form to the lung is 0.22 rem following the inhalation of 1 microcurie. In the soluble form the dose delivered to the bone following the inhalation of 1 microcurie of zirconium-95 is 0.06 rem. The dose delivered to the bone following injection of 1 microcurie into the body via a wound is 0.22 rem. For niobium-95 the above doses are reduced by a factor of three to four.

Values of the maximum permissible total body burden of zirconium-95 and niobium-95 which are deposited in the total body and produce the maximum permissible dose rate to a particular body organ have been compiled.[0563] For both isotopes the critical organ is the bone and the maximum permissible body burden is 20 microcuries for zirconium-95 and 40 microcuries for niobium-95.

3. OTHER HAZARDS

Besides its radiation hazard, zirconium-95 is not significantly poisonous, and so far as is known, the inherent toxicity of zirconium compounds is low.[0776] The toxicity of niobium is unknown.[0776] Zirconium fire and explosive hazard is high, especially in the form of dust when exposed to chemical reaction with air or oxidizing agents. The fire and explosive hazard of niobium is low.

TABLE 5

RADIATION PROTECTION STANDARDS FOR INDIVIDUALS AND POPULATION GROUPS FOR EXTERNAL AND INTERNAL EXPOSURE[2149]

Type of Exposure	Dose to Individuals at Points of Maximum Probably Exposure (rem per year)	Average Dose to a Suitable Population Sample (rem per year)
Whole body, gonads, or bone marrow	0.5	0.17
Thyroid or bone	1.5	0.5
Bone (alternate standards)	Body burden at 0.003 micrograms of radium 226 or its biological equivalent	Body burden of 0.001 micrograms of radium 226 or its biological equivalent

4. DEFINITION OF ADEQUATE WASTE MANAGEMENT

Handling, Storage, and Transportation

Since zirconium-95 and niobium-95 are hazardous to man by inhalation, ingestion, or direct radiation exposure, care is exercised in their handling. Special procedures and radiation shielding are utilized in their handling. The beta particles emitted by them generally cannot penetrate more than 0.1 in. of water or 0.04 in. of glass. Their gamma rays are highly penetrating. Lead and concrete are commonly-used shielding materials. To detect and control personnel exposure, all persons working with these materials should wear dosimetry devices which directly indicate the dose. Commonly used devices are the film badge and the thermoluminescent dosimeters (TLD).

Zirconium-95 and niobium-95 are stored in controlled areas in specially-constructed containers. They are protected by both a primary and a secondary containment barrier. Special monitoring systems and proper warning signs are located in the general area of the storage facility.

Zirconium-95 is classified as a transport group III and niobium-95 as a transport group IV radionuclide by the Department of Transportation, and the rules and regulations governing their transportation are given in the Code of Federal Regulations (CFR) Title 49--Transportation, Parts 170 to 190.[2150] The zirconium-95 content is limited to 3 curies for a Type A package and 200 curies for a Type B package defined in 49CFR173. For niobium-95 the limits are 20 curies for a Type A package and 200 curies for a Type B package. The limits are increased to 20 and 5,000 curies if their physical form meets the requirements of a special form material. Their release rate is limited to zero under the specified accident conditions for Type A and B quantities.[2150] The allowable release of radioactivity from packages containing large quantities of these isotopes is limited to gases and contaminated coolant containing total radioactivity exceeding neither 0.1 percent of the total radioactivity of the package nor 10 curies under the hypothetical accident conditions prescribed in 49CFR173.

Disposal/Reuse

The disposal of zirconium-95 and niobium-95 is governed by the AEC Manual Chapter 0524[0559] and 10CFR20.[2149] Two sets of standards have been established for the permissible radiation exposure in unrestricted areas. One is for the greatest dose received by an individual and the other for the average dose received by the general population. The radiation protection standards for these two groups is attached (Table 5). The standards for the safe disposal of zirconium-95 and niobium-95 also define their maximum concentrations in air and water. Their concentrations in an unrestricted area should not exceed the concentrations in microcuries per milliliter included in this report (Table 6).

Although rarely practiced, the disposal of zirconium-95 and niobium-95 into a sanitary sewage system is limited to 100 microcuries. Their disposal by burial in the soil at any one location and time is limited to 10,000 microcuries.[2149]

5. EVALUATION OF WASTE MANAGEMENT PRACTICES

Recovery

Option No.1 - Scavenging-Precipitation Foam Separation. The removal of zirconium-95 and niobium-95 from low-level radioactive waste water by scavenging-precipitation foam separation has been studied by Oak Ridge National Laboratory.[0703] The process consists of two steps:
(1) precipitating in a sludge-blanket clarification step; and
(2) achieving final decontamination in a foam-separation column.
Processing rates of 300 gal. per hour (gph) for scavenging precipitation and 120 gph for foam separation have been achieved. Decontamination factors (ratio of initial to final concentration)

Table 6

ZIRCONIUM-95 AND NIOBIUM-95 MAXIMUM PERMISSIBLE CONCENTRATIONS[2149]

Isotope	Exposure Group	Form	Concentration in Air microcuries/milliliter	Concentration in Water microcuries/milliliter
Zirconium-95	Individual	Soluble	4×10^{-9}	6×10^{-5}
Zirconium-95	Individual	Insoluble	1×10^{-9}	6×10^{-5}
Zirconium-95	Population	Soluble	1.3×10^{-9}	2×10^{-5}
Zirconium-95	Population	Insoluble	$.33 \times 10^{-9}$	2×10^{-5}
Niobium-95	Individual	Soluble	2×10^{-8}	1×10^{-4}
Niobium-95	Individual	Insoluble	3×10^{-9}	1×10^{-4}
Niobium-95	Population	Soluble	$.67 \times 10^{-8}$	$.33 \times 10^{-4}$
Niobium-95	Population	Insoluble	1×10^{-9}	$.33 \times 10^{-4}$

greater than 50 have been obtained for both isotopes.[0703] At the present time, further development is required to reduce operating costs and increase processing rates.

Option No.2 - Scavenging-Precipitation Ion Exchange. In this process final decontamination is obtained by ion exchange columns from the scavenging-precipitation process. The process includes a provision for the recycle of the ion exchange waste to the scavenging-precipitation step. All the removed radionuclides are concentrated in the clarifier sludge. The overall decontamination factors for these isotopes varied from 10 to 150.[0705] The use of ion exchange resins with a scavenging-precipitation step is a fairly simple and efficient means of removing these isotopes from low-level radioactive aqueous wastes.

Option No.3 - Water Recycle. The water recycle process is used for decontaminating radioactive waste water and recycling the purified water for reuse. The process has been demonstrated at the pilot plant scale by Oak Ridge National Laboratory.[0707] The steps in the process include: (1) clarification by the addition of coagulants; (2) demineralization by cation-anion exchange; and (3) sorption on granular activated carbon. Overall decontamination factors of 350 were obtained. This is the minimum value since the concentrations of zirconium-95 and niobium-95 were reduced to the analytical limits of detection (background). This method is an improvement to the treat-and-discharge methods, but further work is required until full-scale production use is obtained.

Storage/Disposal

Option No. 1 - Land Burial. Land burial of low-level zirconium-95 and niobium-95 wastes in small concentrations at approved sites meeting the geologic and hydrologic criteria, is an acceptable means of disposal. Their concentrations should not be in excess of 10^4 times their maximum permissible concentrations for the general population in 10CFR20.[2149] All wastes to be disposed of should be in a solid form and encased in a suitable container. Liquid wastes should be solidified, preferably using asphalt, in accordance with the methods described in the Radioactive Waste Solidification report. The burial trenches should be designed not to intercept the ground water table and constructed with a bottom drain and sump for water monitoring. The trenches should be covered with either asphalt or vegetation to limit infiltration of water. The burial site design, geology, and hydrology should be in conformance with the criteria used in selecting and licensing the present commercial burial sites.[1423] Since land burial has successfully been practiced on a commercial scale and both zirconium-95 and niobium-95 have short half-lives, it should be considered as the most satisfactory method of disposing of all dilute concentrations of these wastes.

Option No. 2 - Near-Surface Liquid Storage. Near-surface storage of reactor-produced aqueous solutions containing zirconium-95 and niobium-95 high-level waste in carbon steel and stainless steel tanks encased in concrete and buried underground is a satisfactory means of short-term storage. Aqueous solutions of high-level waste have been stored in this manner over the past 25 years. The tanks range in size from 0.33 to 1.3 million gal. and are generally equipped with devices for measuring temperatures, liquid levels, and leaks. These tanks are considered as an interim storage technique due to a general lack of confidence in their long-term integrity.[0705] Since present regulations require the solidification of all reactor wastes within 5 years following reprocessing,[2149] the near-surface storage of aqueous solutions of zirconium-95 and niobium-95 and other fission product salts in steel tanks should only be considered as a near-term storage technique and not as a permanent storage or disposal technique.

Option No. 3 - Near-Surface Solid Storage. The storage of high-level solidified zirconium-95 and niobium-95 wastes and other waste salts in engineered storage facilities offers the best intermediate method for storage of these wastes. The technology for these facilities has been developed. The wastes will be under surveillance and control and can be retrieved, should this be required. The high-level wastes from spent fuel processing should be solidified and packaged in a suitable container. Of the four high-level solidification processes developed, spray or phosphate glass solidification processes offer the better solidified waste characteristics than the pot calcination or the fluidized bed calcination processes (see Radioactive Waste Solidification report). The wastes will be stored in stainless steel-lined concrete vaults which will be air or water cooled. The periodic replacement of the waste containers probably will not be required since the activity of zirconium-95 is reduced by a factor of 100 in 432 days and niobium-95 activity is reduced by a factor of 100 in 232 days. The replacement of the containers may be required if other long-lived isotopes are present.

Option No. 4 - Salt Deposits. This method offers the best potential for the disposal of wastes from fuel reprocessing since bedded salt deposits are completely free of circulating ground waters. This method of disposal has been under study by the Oak Ridge National Laboratory since 1957, and in November of 1970 a committee of the National Academy of Sciences recommended that the use of bedded salt for the disposal of radioactive wastes is satisfactory.[0733] Recent questions concerning the adequacy of this method have resulted in the need for further development work before it can be accepted as an ultimate method of disposal. The wastes must be solidified and packaged in a suitable container. The solidified wastes are buried in rooms carved in the salt deposits, approximately 1,000 ft below the ground. The salt is a good heat transmitter, provides about the same radioactive shielding as concrete, and can heal its own fractures by plastic flow. The critical problem is the selection of a site that meets the necessary design and geological criteria for the mixture of fission product and actinide wastes.

Option No. 5 - Bedrock Disposal. The disposal of high-level aqueous solutions containing zirconium-95 and niobium-95, along with other wastes from spent fuel reprocessing wastes in vaults excavated in crystalline rock over 1,500 ft beneath the ground is currently being evaluated by E. I. du Pont de Nemours, at their Savannah River Plant near Aiken, South Carolina.[0894,1396] The wastes would be stored in six tunnels and once in the tunnels the wastes will seep into the surrounding rock. Located above the crystalline rock is the Tuscaloosa formation, a good source of freshwater, which is separated by a layer of clay that would act as a barrier to the leakage of radioactive wastes. An advisory committee appointed by the National Academy of Sciences recommended abandonment of the project. In May 1972, another National Academy of Sciences panel concluded that bedrock storage provides a reasonable prospect for long-term safe storage, but precise information is needed to decide if and where underground storage vaults should be built. Another method has also been proposed for disposing of liquid wastes by in situ incorporation in molten silicate rock.[2146] At the present time both of these methods are unproven since sufficient engineering data or exploration has not been completed to verify their suitability.

Option No. 6 - Hydraulic Fracturing. The direct disposal of aqueous low-level radioactive wastes into shale formations has been investigated by Oak Ridge National Laboratory.[0709] The method consists of mixing the aqueous wastes with cement and pumping the resulting slurry down a well out into a nearly horizontal fracture in a thick shale formation. Since zirconium-95 and niobium-95 are leached from cement and no additional encapsulation is provided, this method of disposal requires additional work to determine its suitability.

To summarize, both zirconium-95 and niobium-95 are short-lived isotopes with a high initial activity but whose activity decreases by a factor of 1,000 within 2 years. These isotopes will probably be contained with the long-lived fuel reprocessing wastes since their recovery is not required due to their short half-life and limited commercial use. The acceptable method of treatment is solidification followed by storage in near-surface engineered facilities and disposal in salt deposits.

6. APPLICABILITY TO NATIONAL DISPOSAL SITES

Zirconium-95 and niobium-95 are both candidates for a National Disposal Site due to their projected growth with that of the civilian nuclear power program. From a cost and safety viewpoint, it would be desirable to combine the reprocessing plant and the National Disposal Site. Interim storage in near-surface engineered facilities offers considerable latitude in the site selection. The site selection for ultimate disposal of these wastes is limited to particular geological areas in which salt deposits are present.

The recommended treatment for high-level aqueous wastes containing zirconium-95 and niobium-95 is solidification using either spray or phosphate glass solidification processes. If required for economic reasons, recovery prior to solidification could be effected by scavenging-precipitation ion exchange. The solidified wastes should then be held in interim storage in engineered storage facilities, and finally disposed of in salt deposits.

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HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Zirconium-95

Half-life 65 days

Type of Decay Negative Beta

Molecular Wt. 95

Melting Pt. 1852C

Boiling Pt. 3578C

Density 6.49 gm/cc

Structural Formula

$_{40}Zr^{95}$

Specific Power 111 watts/gm

Specific Activity 2.12×10^4 curies/gm

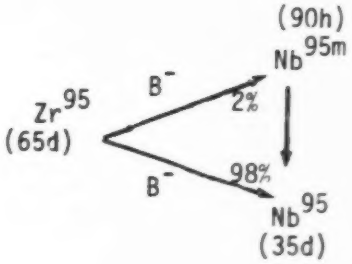
Solubility

Cold Water Insoluble

Hot Water Insoluble

Others: Soluble in HF and aqua regia, slightly soluble in acid

Decay Chain



Radiation Energy Level & Intensities

Beta:	0.99 Mev (2%)
	0.396 Mev (55%)
	0.360 Mev (43%)
Gamma:	0.724 Mev (49%)
	0.756 Mev (49%)

Shipping Regulations Classified as a transport group III radionuclide by the Department of Transportation

Comments

References: (1) 0776
(2) 2150

HAZARDOUS WASTES PROPERTIES
WORKSHEET

Name Niobium-95

Structural Formula $_{41}Nb^{95}$

Half-life 35 days

Type of Decay Negative Beta

Molecular Wt. 95 Melting Pt. 2468C Boiling Pt. 4927C

Density 8.57 gm/cc

Specific Power 190 watts/gm

Specific Activity 3.93×10^4 curies/gm

Solubility

Cold Water Insoluble Hot Water Insoluble

Others: Soluble in fused alkali; insoluble in HCl, HNO_3 and aqua regia

Decay Chain

$$\underset{(35d)}{Nb^{95}} \xrightarrow{B^-} \underset{(stable)}{Mo^{95}}$$

Radiation Energy Level & Intensities

Beta: 0.160 Mev (99%)
0.924 Mev (1%)
Gamma: 0.765 Mev (100%)

Shipping Regulations Classified as a transport group IV radionuclide by the Department of Transportation

Comments

References: (1) 0776
(2) 2150